

A sustainable approach for mitigating rebar corrosion in adverse conditions using *Phyllanthus emblica* (L.) leaf extract as concrete scale inhibitor

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Abstract

The primary challenge of reinforced concrete infrastructures (ReCIs) is the early corrosion damage of their reinforcing mild steel (ReMS). Effective and sustainable approaches are needed to reduce reinforced concrete corrosion (ReCC). This research evaluates the effectiveness of *Phyllanthus emblica* (L.) leaf extract (PEL) as a concrete inhibitor for the first time, aiming to minimize the early corrosion risk of ReMS in three harsh circumstances such as simulated industrial environment (SIdE), simulated coastal environment (SCoE), and ordinary environment (OrdE) of ReCIs. To achieve this, fifteen reinforced concrete cuboid beams (RC2Bs) were formulated using concrete mixes with varying PEL concentrations (500 ppm, 1000 ppm, 2000 ppm, and 4000 ppm) in these three harsh electrolytes. The study measured the corrosion potential (E_{cor}) and electrochemical resistivity ($EChR$) of the ReMS rod in the RC2Bs at regular intervals until seven months. The measurement of the electrochemical parameters E_{cor} and $EChR$ has shown an effective, sustainable, and cost-efficient method for monitoring the condition of ReCC in real-time and on-site, following the ASTM C876-22b. Screening tests, including UV-Vis spectroscopy, FTIR, and GC-MS/UHPLC-MS, confirmed the presence of secondary metabolites (SMBs) in PEL, which act as anti-corrosive agents to help prevent early corrosion of reinforcing steel in concrete beams. The E_{cor} value improved to -125 mV (SCE) or more positive potential, and the $EChR$ increased by more than $20 \text{ k}\Omega \cdot \text{cm}$ with high PEL concentrations after seven months of exposure, indicating a lower corrosion risk of the ReMS. Surface analyses of the ReMS rods were conducted before and after reinforcement in the RC2Bs, validating the electrochemical findings.

Received: May 27, 2025. Published: September 13, 2025

doi: [10.17675/2305-6894-2025-14-3-26](https://doi.org/10.17675/2305-6894-2025-14-3-26)

Keywords: effective inhibitor, emblica leaf extract, marine and industrial conditions, SEM-EDS, rebar coatings.

List of abbreviations

Abbreviation	Full name
ASTM	American Society for Testing Materials
E_{cor}	Corrosion potential
<i>EChR</i>	Electrochemical resistivity
FTIR	Fourier transform infrared
GC-MS	Gas chromatography mass spectrometry
HCorZ	High corrosion risk zone
ICorZ	Intermediate corrosion risk zone
LCorZ	Low corrosion risk zone
MilS	Mild steel
OM	Optical microscopy
OrdE	Ordinary environment
OPC	Ordinary Portland cement
PEL	<i>Phyllanthus emblica</i> (L.) leaves extract
RC2B(s)	Reinforced concrete cuboid beam(s)
ReCC	Reinforced concrete corrosion
ReCI(s)	Reinforced concrete infrastructure(s)
ReMS	Reinforcing mild steel
SCE	Saturated calomel electrode
SCoE	Simulated coastal environment
SEM-EDS	Scanning microscopy with energy dispersive X-ray spectroscopy
SIde	Simulated industrial environment
SMB(s)	Secondary metabolites
3D	Three dimensional
2D	Two dimensional
UHPLC	Ultra-high performance liquid chromatography
UV-Vis	Ultraviolet visible spectrometry
WLI	White light interferometer

1. Introduction

The proliferation of concrete usage is a direct outcome of rapid urban development and industrial expansion. Reinforced concrete is the second most widely used construction material globally, following water [1]. The widespread application of reinforced concrete has demonstrated high mechanical properties, tensile strength, and resilience [2]. The ReCIs are resilient, long-lasting for fifty years or more, and are commonly used in industrial operations [3]. However, reinforcing mild steel (ReMS) of the RCIs is prone to corrosion and can quickly deteriorate and weaken RCIs when exposed to corrosive environments [4]. Extended deterioration of the ReMS can cause structural instability, making the concrete structures unable to support themselves. The inactive or corrosion-protective nature of the ReMS rod in concrete beams is due to the formation of a very thin passivating nano-layer that develops at the interface regions between the concrete mix and the ReMS surface at a pH range between 12.5 and 13.6 [5]. Entry of atmospheric acid ion pollutants, such as sulfates, sulfides, and CO₂, causes drops of ReCIs pH 9–10 or below. They caused the passive layer to destabilize, which in turn caused reinforced concrete damage and the eventual collapse of the ReCIs, as described in the mechanisms of sulfation/sulfidation and carbonation elsewhere [6].

The methods of ReCC control have a range of practices [7, 8], including the admixture of various corrosion inhibitors in the concrete mix [9]. Among these, inorganic, organic, and hybrid inhibitors managed the corrosion control of ReCIs in early periods. These inhibitors are cathodic, anodic, or mixed based on the active inhibitor molecules that help to minimize the electrochemical corrosion process, the formation of thin barrier films, physico-sorption, and chemisorption [10]. In the last few decades, phytochemicals having heteroatoms and aromatic ring systems, such as alkaloids, flavonoids, terpenoids, and phenolic compounds, found in plant extracts, are typically derived from leaves, bark, stem, seeds, and flowers, are proven to be effective for minimizing the corrosion of various metals in aggressive electrolytes even in concrete solution. Upon the addition of these plant-based phyto-molecules as corrosion inhibitors to the concrete mixture, they adsorbed onto the reinforcing metal's surface by the process of their hydrolysis, donor-acceptor linkage between the metal and π -electrons, or unshared electrons and the empty d-orbitals of the Fe-atoms, and vice versa [11].

Over the past decade, there has been significant analysis of the anti-corrosion properties of various phyto-extracts derived from the leaves of plants native to Nepal. These include floss-flower and yam [12], sesame [13], mango [14], guava [15], the five-leaved chaste tree and Madagascar periwinkle [16], heart-leaved moonseed [17], and acacia and sweet bay [18]. These plant extracts exhibit enhanced corrosion-inhibiting properties across a wide range of electrolytic environments, from aqueous solutions. Different anti-corrosive compounds, particularly plant-derived phyto-molecules, can be incorporated into concrete mixes to reduce specific forms of ReCC. In this context, we have taken the example of an

anti-corrosion concrete admixture, considering the inhibiting effect of the methanolic extract of *Phyllanthus emblica* (*Amalaa* in Nepali) leaves.

Phyllanthus emblica Linn, a well-known member of the *Euphorbiaceae* family, is extensively spread in several tropical and subtropical nations [19]. A phytochemical screening test of methanolic *P. emblica* leaf extract previously confirmed the presence of phyto-compounds like isopropyl palmitate, linolenic acid, 1,5-hexanediol, and 1,3-dimethyl indole from the GC-MS analysis [20]. Furthermore, limited research has explored the application of this plant's bark extract as an anti-corrosive agent for the corrosion control of ReCIs in aggressive environments [21]. In light of these considerations, this study extracted bioactive phyto-compounds from the leaves of the *Phyllanthus emblica* (L.) plant. To the author's knowledge, phyto-compounds derived from the methanolic leaf extract of the *Phyllanthus emblica* (L.) plant exploit for the first to control the corrosion of ReCIs in two simulated conditions, simulated coastal environments (SCoE) and simulated industrial environments (SIde), including ordinary environments (OrdE).

The research determines how methanolic leaf extract inhibits the corrosion of ReMS. Also, the study ascertains whether the *Phyllanthus emblica* (L.) leaf extract effectively prevents corrosion by conducting an *in situ* electrochemical analysis in compliance with ASTM C876-22b standards [22]. Phytochemical profiling, UV-visible, and FTIR spectroscopic studies characterize the plant extracts. Besides, the anti-corrosion behavior of the suggested inhibitor on the ReMS rod works out using surface imaging methods. A future goal would be to create green and eco-friendly corrosion inhibitors as concrete admixtures on an industrial scale.

2. Materials and Methods

2.1. Preparation of *Phyllanthus emblica* leaf extract

The branches with fruit of *Phyllanthus emblica* (L.), which is one of the widely distributed Nepali plants, were collected from Siddarthagar-8, located in the Rupendehi district of Nepal, at an altitude of 109 meters above sea level (coordinates: 27.5207°N, 84.4437°E). The location map of the plant collecting site is in Figure 1. The plant specimen was submitted to the National Herbarium and Plant Laboratories at the Department of Plant Resources in Godawari, Lalitpur, Nepal, for authentication of its botanical name.

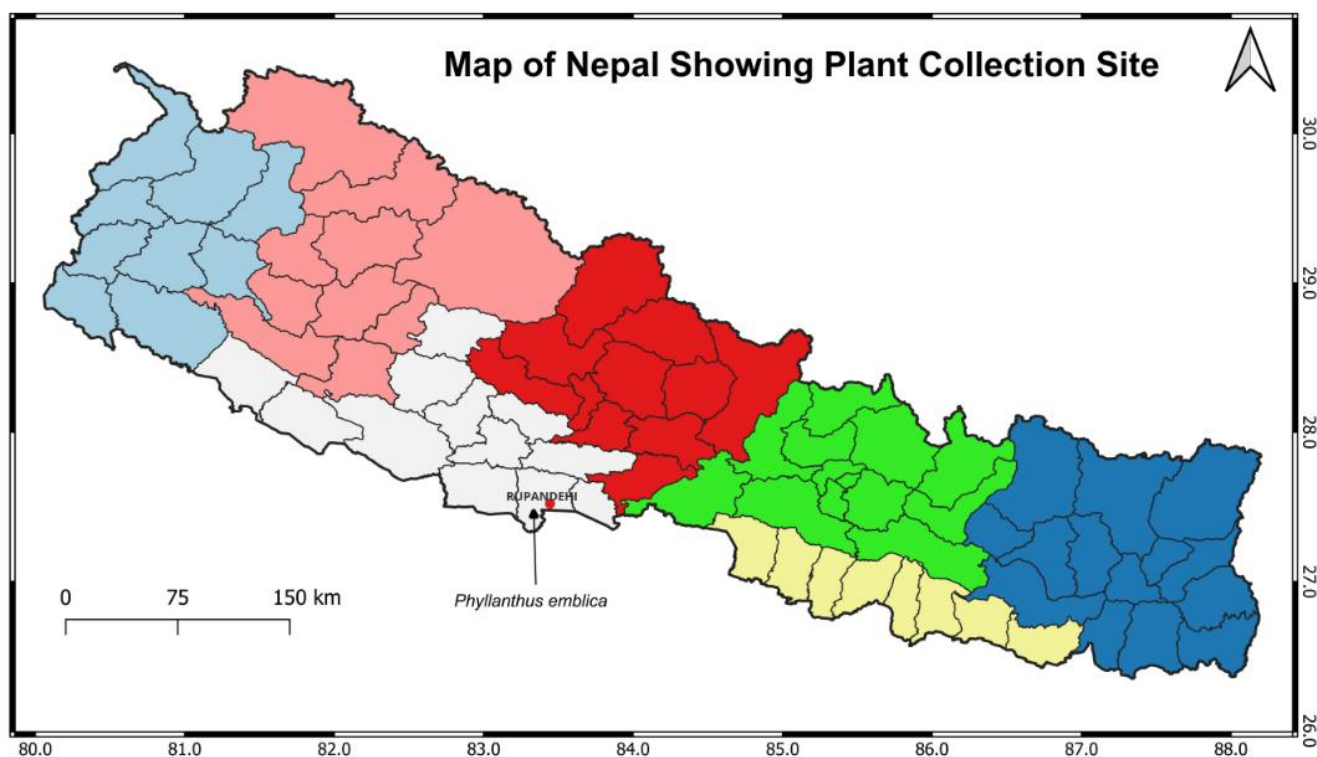


Figure 1. Geographical map of the *Phyllanthus emblica* (L.) leaves collection site.

About 1000 grams of the collected leaves of the *Phyllanthus emblica* L. (Figure 2(a)) were washed with distilled water, and dried in the shaded areas. The dried leaves were ground into a powder (Figure 2(b)) using an electric grinder and soaking 400 g ground powder in 800 mL methanol at a 1:2 ratio for two weeks (Figure 2(c)) using a process of percolation (Figure 2(d)), as described in Ref. [23]. After the percolation, the mixture was separated into filtrate and residue using Whatmann®Grade-1 (UK) filter paper, as depicted in Figure 2(e), and subjected to concentration (Figure 2(f)) the filtrate in a rotary evaporator (IKA®RV 10 digital V, Germany) at 40°C to procure a semi-solid leaf extract of the *Phyllanthus emblica* (L.) plant, abbreviated as PEL. The concentrated semi-solid PEL was stored at 4°C or below temperature before its utilization as a concrete admixture and characterized using different chemical screening tests, spectroscopy, and chromatographic analyses, as explained elsewhere [21].

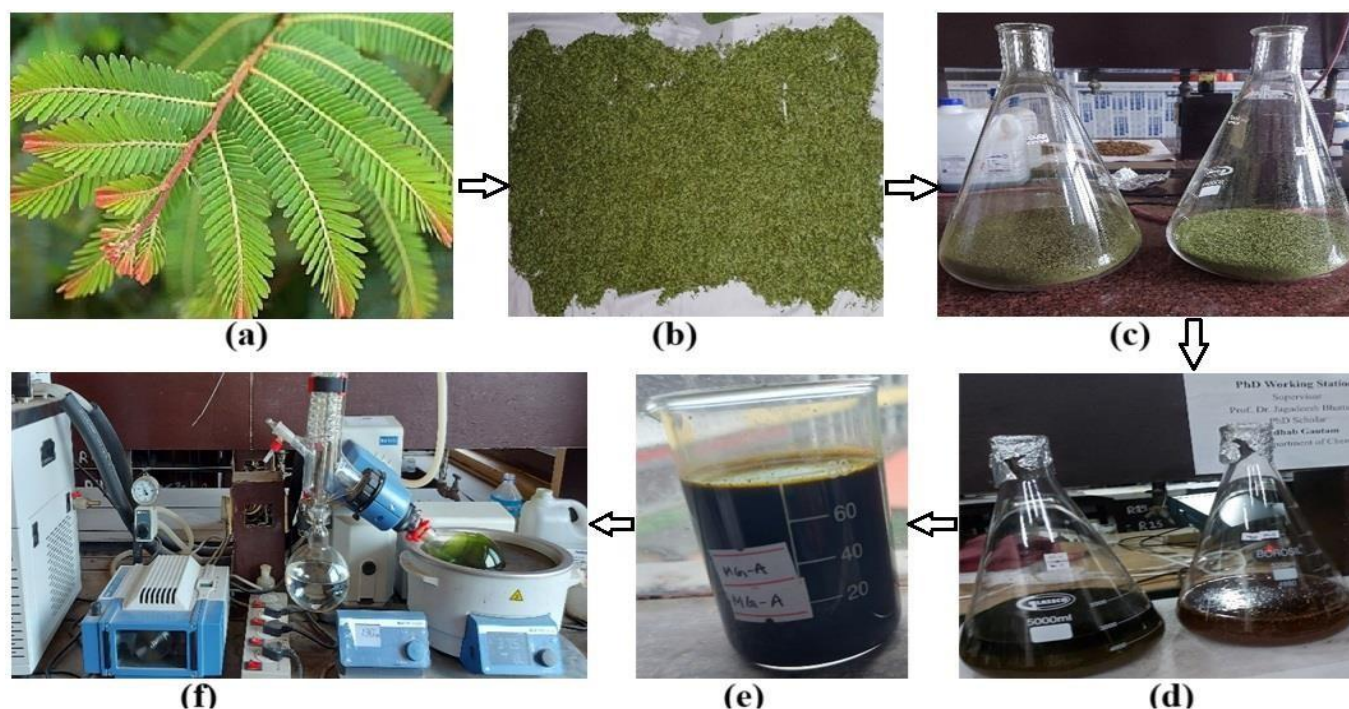


Figure 2. Steps of PEL preparation; fresh leaves of *P. emblica* (a), dried leaf powder of *P. emblica* (b), leaf powder soaked in methanol (c & d), phyto-molecules consisting methanolic filtrate (e), and concentration of the filtrate in a rotary evaporator (f).

2.2. Preparation of reinforced concrete beam

Gathered OPC cement (Grade@53) powder, impurity-free fine sand (river), gravel (20 mm diameter), and distilled water from the local market of the Kathmandu Valley to collect the concrete aggregates for the formulation of reinforced concrete beams, as described elsewhere [14]. Five concentrations (*i.e.*, 0, 500, 1000, 2000 and 4000 ppm) of PEL extract in SCoE (0.5 M NaCl), SiDE (0.5 M Na₂SO₄), and OrDE (without NaCl/Na₂SO₄) mixed with required amounts of concrete aggregates having a ratio of 1:2:3 ratio cement, sand, and gravel, and 0.45 (cement/water) ratio to prepare the concrete paste (Figure 3(a)), as previously established protocol [15].

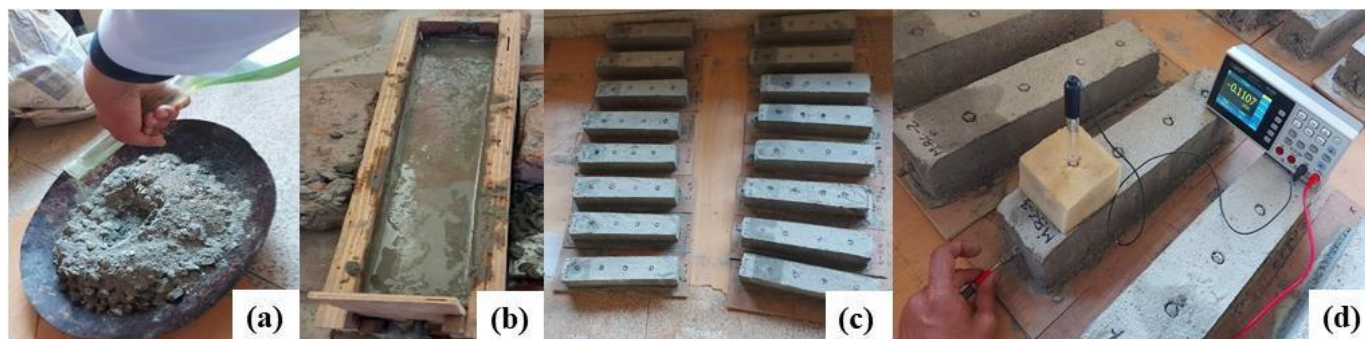


Figure 3. (a) Cement-sand-gravel-PEL for preparation of RC2Bs, (b) casting in cuboid molding box, (c) prepared RC2Bs exposed in outdoor, and (d) measurement of E_{cor} and E_{ChR} using bench type multi-meter.

We abraded the collected mild steel (MilS) rod from the local market using abrasive paper (grit numbers 600 to 2000) until all surfaces of the sample specimens were polished to a mirror finish, removing any rough edges. Additionally, the polished samples were cleaned using ultrasonic waves before being used to fabricate the reinforced concrete samples. Some specimens were stored in a desiccator until used. Aqueous solutions of 0.5 M NaCl, 0.5 M Na₂SO₄, and without both salts, abbreviated as the SCoE, SIde, and OrdE electrolytes, respectively, were used in this investigation. Fifteen rust-free MilS rods (12 mm diameter) were fixed at a central point of a rectangular box of (40×8×8) cm³, as illustrated in Figure 3(b), to cast 15 RC2Bs (summarized in Table 1). Following the fabrication of the RC2Bs, they were cured for one week on the laboratory balcony under dry atmospheric conditions.

Table 1. Compositional details of fifteen reinforced concrete cuboid beams (RC2Bs) using concrete paste with PEL and NaCl/Na₂SO₄.

S. No.	Sample Name	Composition of RC2Bs
1	0 ppm PEL (Control)	Control
2	500 ppm PEL	Concrete paste with 500 ppm PEL
3	1000 ppm PEL	Concrete paste with 1000 ppm PEL
4	2000 ppm PEL	Concrete paste with 2000 ppm PEL
5	4000 ppm PEL	Concrete paste with 4000 ppm PEL
6	0 ppm PEL-SCoE	Control SCoE
7	500 ppm PEL-SCoE	Concrete paste with 500 ppm PEL+0.5 M NaCl
8	1000 ppm PEL-SCoE	Concrete paste with 1000 ppm PEL+0.5 M NaCl
9	2000 ppm PEL-SCoE	Concrete paste with 2000 ppm PEL+0.5 M NaCl
10	4000 ppm PEL-SCoE	Concrete paste with 4000 ppm PEL+0.5 M NaCl
11	0 ppm PEL-SIde	Control SIde
12	500 ppm PEL-SIde	Concrete paste with 500 ppm PEL+0.5 M Na ₂ SO ₄
13	1000 ppm PEL-SIde	Concrete paste with 1000 ppm PEL+0.5 M Na ₂ SO ₄
14	2000 ppm PEL-SIde	Concrete paste with 2000 ppm PEL+0.5 M Na ₂ SO ₄
15	4000 ppm PEL-SIde	Concrete paste with 4000 ppm PEL+0.5 M Na ₂ SO ₄

2.3. Electrochemical corrosion potential and resistivity measurements

After curing the fifteen RC2Bs, as depicted in Figure 3(c), E_{cor} and $EChR$ were monitored from the 1st to 30th weeks or more exposure using a digital bench-type multi-meter (OWON XDM1241, China). Four points were marked (*i.e.*, 1, 2, 3, 4) in each cuboidal concrete beam surface, and they are almost equidistant from all sides of the beam. For the electrochemical

analysis, the exposed end of the ReMS rod serves as the working electrode. Besides, a saturated calomel electrode (SCE) as a reference electrode that is inserted into the wetted sponge by aqueous surfactant, as in Figure 3(d), is placed just over the marked surface points of the beams obeying the method described elsewhere [24].

The E_{cor} and $EChR$ values for all four points of each RC2B were recorded at 7, 14, 21, ..., and up to 210 days or more and repeated the procedure for all sets of RC2Bs. The text outlines a methodology for predicting the corrosion risk levels associated with ReMS rods by utilizing recorded electrochemical properties, the E_{cor} , and $EChR$ values detailed in Table 2.

Table 2. Qualitative prediction of the corrosion probability of the ReMS in concrete, based on E_{cor} and $EChR$.

E_{cor} (mV vs SCE)	$EChR$ ($\text{k}\Omega \cdot \text{cm}$)	Corrosion risk condition of RC2B
< -276	< 10	High or $> 90\%$ – Corrosion Risk Zone (HCorZ)
-275 to -126	10 to 20	Intermediate or 10 – 90% – Corrosion Risk Zone (ICorZ)
> -126	> 20	Low or Less than 10% – Corrosion Risk Zone (LCorZ)

The findings reveal that a E_{cor} value of less than -276 mV (SCE) or an $EChR$ value below $10 \text{ k}\Omega \cdot \text{cm}$ indicates that the ReMS rod resides within the high-corrosion risk zone. Conversely, a E_{cor} value exceeding -126 mV (SCE) along with an $EChR$ value greater than $20 \text{ k}\Omega \cdot \text{cm}$ classifies the ReMS rod within the low-corrosion risk zone. In cases where the E_{cor} and $EChR$ values are intermediate, the corrosion risk is intermediate. For instance, if the E_{cor} of a ReMS rod is at -289 mV (SCE) and the $EChR$ is recorded at $13 \text{ k}\Omega \cdot \text{cm}$, such that the ReMS rod would be deemed to fall within the intermediate corrosion risk zone.

2.4. Characterization of materials

The materials of PEL extract, OPC cement, and MilS were identified using techniques of UV-vis (SPECORD®200PLUS, Germany), FTIR (IR Affinity-1S, SHIMADZU), and a GC-MS (an Agilent 7890B GC apparatus coupled with a 5977A Mass Selective Detector) spectrometric techniques to analyze the functional groups and number of phyto-molecules, respectively, in the PEL extract. An ultra-high-performance liquid chromatography (UHPLC) technique was applied to confirm the molecular structures of the phyto-molecules present in the PEL, using Xevo G2-XS QToF (Waters Corporation., USA) at Central Drug Research Center of CSIR in Lucknow, India. X-ray diffraction technique (Ultima IV, Japan) at 5 – 80 degrees with a scanning velocity of 4 degrees/min was used to characterize the OPC cement and MilS phases.

2.5. Surface analysis of reinforcing mild steel

The optical microscopic (OM) and scanning microscopic (SEM) images of the ReMS rod surfaces before and after reinforcement in RC2Bs were analyzed using stereomicroscopy

(Olympus SZ 61, Japan) and scanning electron microscopy (SEM, JEOL-USA). The surface composition of the passive layer formed on the ReMS rod surface was analyzed using the energy-dispersive X-ray spectroscopy (EDS) techniques. Furthermore, the ReMS specimens were subjected to analysis to capture three-dimensional (3D) surface images and evaluate average surface roughness and root mean square roughness, employing a white light interferometer (NewView-9000, Zygo Corporation, USA).

3. Results and Discussion

3.1. Chemical profiling test

Different phytochemical tests pinpoint the distinct secondary metabolites (SMBs) existing in the PEL extract, as documented in a prior study [11]. The results of the tests on PEL verified the existence of alkaloids, flavonoids, phenols, saponins, steroids, terpenes, and glycosides, as outlined in Table 3. The phytochemical test results showed that the PEL contains a more diverse range and higher amounts of SMBs than in bark extract of the same plant species in methanol solvent (for example, the saponin is absent in the bark extract of the *Phyllanthus emblica* (L.), as reported in the literature [21]. The screening results indicate the presence of saponin in the PEL, consistent with findings reported in previous studies on the ethanol leaf extract of *Phyllanthus emblica* [25]. Earlier research indicated that green extract effectively serves as an anti-corrosive agent in concrete mixture and forms a protective layer on the surface of the ReMS rod, thereby preventing ReCC ReCC [26]. These SMBs possess unsaturated π -bond systems, aromatic ring configurations, and heteroatoms such as N, O, and P, and they exhibit a high affinity for adsorption onto the ReMS surface through chemical and physical adsorption mechanisms.

Table 3. Phytochemical profiling tests on PEL.

SMBs	Tests	PEL
Alkaloids	Dragendroff's Test	+++
Flavonoids	Alkaline Test	++
Phenolics	Lead Acetate Test	++
Saponins	Foam Test	+
Tannins	Ferric Chloride Test	+
Terpenoids	Salkowski Test	+
Steroids	Liebermann-Burchard Test	++
Glycosides	Legal Test	+

–=absence, +=presence & ++/+++ =presence of high concentration

3.2. Spectral analyses of PEL

The UV-Vis and FTIR spectral analyses of PEL verified the presence of different SMBs. Various UV-Vis peaks are observed in PEL, particularly between 200 and 800 nm, as shown in Figure 4(a). The appearance of eight UV-Vis peaks at 355, 363, 410, 434, 472, 534, 607, and 664 nm in the UV-Vis spectra affirmed the presence of different phytoconstituents in PEL. Previous research has shown that SMBs containing σ -bonds, π -bonds, lone pairs of electrons, chromophores, and aromatic ring compounds exhibit characteristic peaks at around 350 nm and 664 nm [27]. In previous studies, the UV-Vis peaks at 409 nm and 472 nm were identified as tannins and terpenoids [28], respectively, occurring between 350 and 500 nm [29]. Peaks at 410 nm, 472 nm, and 534 nm are characteristic of terpenoids, as reported in the literature [30]. Peaks at 607 nm and 664 nm are identified as chlorophyll [29]. However, these eight UV-Vis peaks for the PEL extract have not been reported, except for one peak at 355 nm in the methanol *Phyllanthus emblica* (L.) bark extract [21].

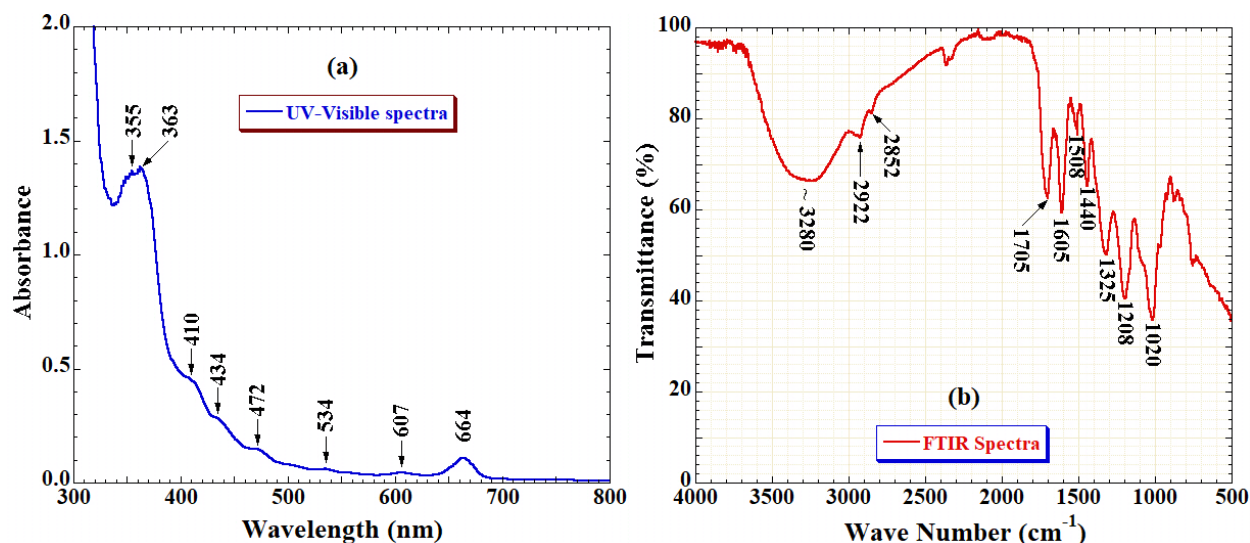


Figure 4. UV-visible (a) and FTIR spectra (b) of PEL for analysis.

Moreover, FTIR analysis facilitates the qualitative identification of functional groups of the PEL extract. Figure 4(b) illustrates the FTIR spectra recorded for PEL. The functional groups show distinct peaks at specific wavelengths: approximately 3280 cm⁻¹, which corresponds to O–H or N–H stretching vibrations in aromatic compounds [31]; 1605 cm⁻¹, indicating aromatic C=C stretching [32]; 1440 cm⁻¹ for C–H stretching [33]; 1208 cm⁻¹ for the –COOH group [34]; and 1020 cm⁻¹ for O–H or C–H bending in aromatic structures [35]. These peaks suggest the presence of phenolic or alcoholic compounds [36]. Furthermore, a peak centered at 2922 cm⁻¹ denotes the asymmetric stretching of C–H vibrations within the methanolic leaf extracts of the plant. These findings are consistent with previous FTIR analysis on the bark extract of this plant [21], except FTIR peaks at 2852 cm⁻¹, 1705 cm⁻¹, and 1508 cm⁻¹, as demonstrated in Figure 4(b). Therefore, the PEL contains a variety of phytoconstituents like alcohols, amines, ketone acid, and unsaturated

compounds, which can build a barrier layer across the surface of the ReMS rod, preventing hostile ions from interacting with Fe and causing corrosion.

A qualitative analysis of the crude methanolic PEL was performed using GC-MS analysis with an Agilent 7890B gas chromatography system connected to a 5977A mass selective detector. This analysis identified approximately 25 different types of SMBs in the PEL extract, as shown in the GC spectra of PEL (see Figure 5). However, the ethanol extract of *Phyllanthus emblica* (L.) leaves reported twenty-two SMBs in the literature [37]. Notably, the present study demonstrated that the methanol PEL extract has a higher abundance of SMBs than the methanol bark extract of the same plant, which constituted only 17 peaks, as reported in a previous study [21].

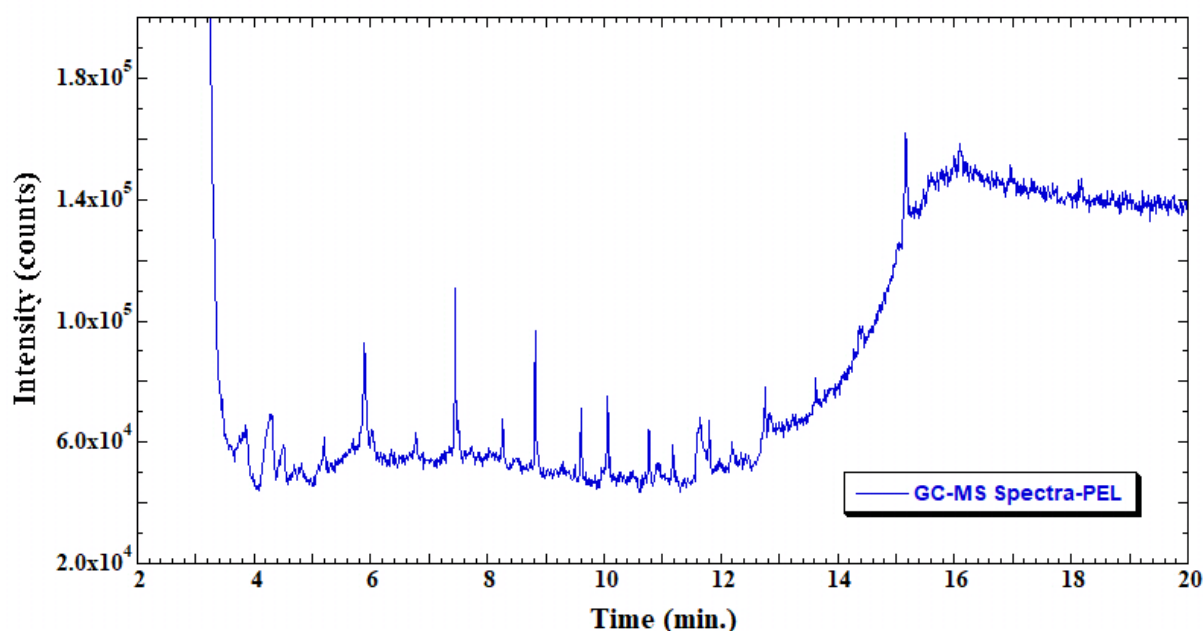


Figure 5. GC spectra of PEL.

Therefore, it presumes that the present studied methanol PEL extract should have better anti-corrosion properties than those of the ethanol leaf extract and the methanol bark extract of the *Phyllanthus emblica* (L.) plant. Hence, this study aims to demonstrate that PEL should have better anti-corrosive properties than the bark extract of the same plant species, contributing to the corrosion reduction of ReMS used in RC2Bs. A qualitative analysis of the crude methanolic PEL was performed using GC-MS with an Agilent 7890B gas chromatography system connected to a 5977A mass selective detector. This analysis identified approximately 25 different types of SMBs present in the PEL extract, as shown in the GC spectra of PEL (see Figure 5).

In addition, the UHPLC technique was employed to validate the molecular structures of the SMBs present in the PEL. This analysis identified thirteen potential phyto-molecules and confirmed their structural formulas and molecular masses through the Mass-Bank search database (<https://massbank.eu/MassBank/Search>). They are 5-(4-bromophenyl)-1-(2,4-

dichlorophenyl)-4-ethyl-*N*-piperidin-1-ylpyrazole-3-carboxamide, α -carotene-4-one, 1,5-dimethyl-2-phenylpyrazol-3-one, quercitrin, kaempferol, fumonisins B3, 5(*S*)-5-carboxy strictosidine, aminophylline, 2',6'-dihydroxy-4-methoxychalcone-4'-*O*-neohesperid, azadirachtin, chloridazon, and cycloxydim sulfoxide. Among these phyto-molecules, polyphenolic flavonoid compounds like quercitrin and kaempferol exhibited potent antioxidant activity [38]. Figure 6 illustrates some structural formulas of the most abundant phyto-molecules revealed in the PEL extract.

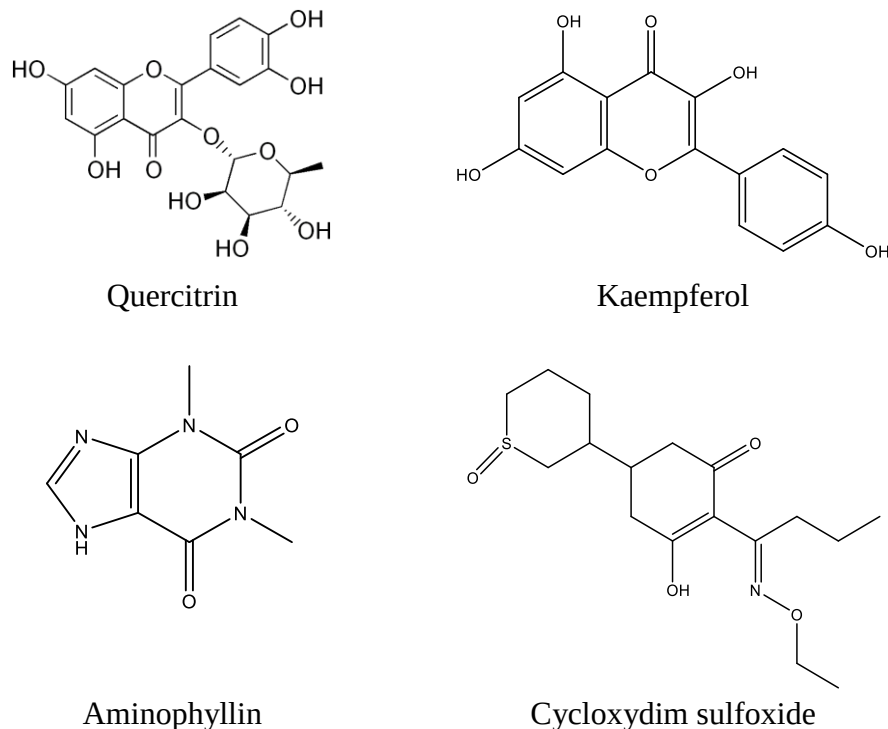


Figure 6. Structural formulas of some secondary metabolites of PEL extract.

Consequently, physical or chemical adsorption and interactions are responsible for the adsorption. Adsorption and interaction processes are likely enhanced by the heteroatoms and unsaturated aromatic systems, as demonstrated by UV-Vis, FTIR, and UHPLC studies in this investigation. These findings emphasize the significant potential of PEL as an anti-corrosive agent to prevent the ReCC. Furthermore, they offer valuable insights into the adsorption mechanisms that facilitate the formation of a protective film on the surface of corroded ReMS, as elaborated upon in additional references [39].

3.3. XRD analysis of mild steel and OPC cement

X-ray diffraction (XRD) analysis is essential for examining the characteristics of building materials like bricks [40], mild steel [41], cement [42], ceramics, and tiles [43]. It helps identify their components and crystal phases. Ordinary Portland cement (OPC) mainly contains four varieties: alite (C_3S), belite (C_2S), aluminate (C_3A), and aluminoferrite (C_4AF), along with minor components like gypsum and calcium sulfate hemihydrate, which

can exist in various forms [44]. In Figure 7(a), distinguishing peaks are observed at $2\theta=29.40^\circ$ (0.3035 nm) and 47.25° (0.1922 nm), which differentiate the γ -C₂S phase from the β -C₂S phase [45]. The monoclinic phase of alite (C₃S) exhibits a discernible peak at $2\theta=29.40^\circ$ (0.3035 nm), which does not significantly interfere with the β -C₂S phase. Consequently, the peak observed at 0.3035 nm is a practical parameter for precise quantitative examination of the monoclinic alite phase within the OPC cement sample [46]. The XRD peak at 41.23° (0.2188 nm) in the cement powder indicates the presence of the belite β -C₂S phase in the sample. The analysis of the OPC cement revealed a diffraction peak at 51.69° (0.1767 nm), confirming the presence of an alite phase, consistent with findings in the literature [47].

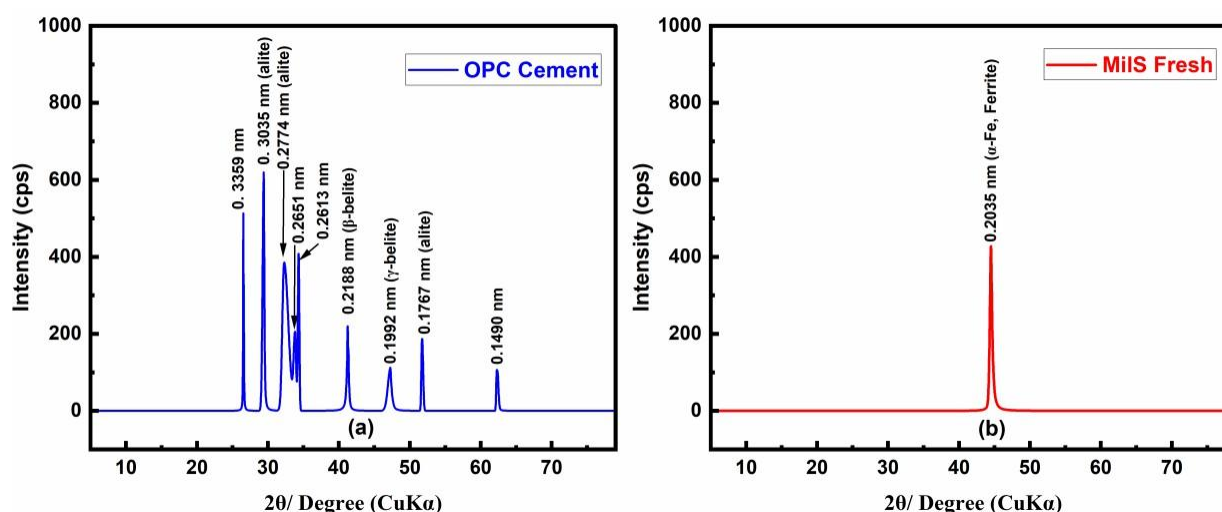


Figure 7. XRD patterns of the (a) OPC cement powder and (b) fresh mild steel surface.

Overall, the alite peaks are identified as a significant phase, indicating it is the main mineral phase of the analyzed OPC cement powder, followed by the belite phase [46]. Similarly, a fresh mild steel (MilS) has a well-defined crystallographic aspect and a long-range periodic atomic structure [41]. Figure 7(b) depicts the XRD patterns of a fresh MilS, which shows only one prominent peak at $2\theta=44.48^\circ$ (0.2035 nm), without other peaks. The data indicate the presence of the α -Fe (ferrite) polycrystalline phase, consistent with findings from prior XRD studies [41].

3.4. Analysis of electrochemical corrosion potential and resistivity

After identifying the materials used to make RC2BS, we estimated the effectiveness of PEL in controlling the early corrosion of ReMS in concrete paste. We used *in situ* and non-destructive electrochemical measurements, specifically the E_{cor} and E_{ChR} methods, to assess three levels of corrosion probability: Low corrosion risk zone (LCorZ), intermediate corrosion risk zone (ICorZ), and high corrosion risk zone (HCorZ) for the ReMS in ReCIs. Based on shifts in E_{cor} and E_{ChR} over exposure durations of up to 210 days, the examination assesses the corrosion possibility levels and the probability of corrosion for the ReMS in a

concrete beam. The RC2Bs manufactured at controlled conditions – OrdE, SCoE, and SIdE – show average E_{cor} values varying from -330 to -30 mV (SCE) during the initial exposure duration of one to eight weeks. Over time, the average E_{cor} in these exposure intervals shifts from high corrosion (HCorZ/ICorZ) to low corrosion (LCorZ). This shift implies a shift in corrosion rates from active to inactive (passive) states, as illustrated in Figure 8(a).

Such observation comes from the alkaline nature of concrete to develop a protective layer on the ReMS surface and inhibit corrosion in aggressive environments. With prolonged exposure, the dissolution of calcium hydroxide leads to a decrease in pH, augmenting the corrosive nature of three control conditions (OrdE, SCoE, and SIdE) towards the reinforcing mild steel, as reported in the literature [47, 48]. Consequently, E_{cor} falls within the intermediate corrosion risk zone (ICorZ), with values ranging from -275 to -125 mV relative to SCE after extended exposure of the RC2Bs fabricated using concrete paste without (OrdE) and with 0.5 M NaCl (SCoE) or 0.5 M Na₂SO₄ (SIdE) electrolyte. When different concentrations (*i.e.*, 500, 1000, 2000, and 4000 ppm) of PELs are added to RC2Bs in OrdE, SCoE, and SIdE, respectively, in Figures 8(b), 8(c), and 8(d). There is a continuous increase in E_{cor} values from ICorP/HCorP to LCoP after 2–3 months. Subsequently, the E_{cor} values of all RC2Bs nearly reach a stable condition, as shown in Figures 8(b), 8(c), and 8(d). Notably, after 210 days of exposure, the E_{cor} consistently exceeded -126 mV relative to the SCE across all three electrolytes.

In OrdE, the ReMS exhibited a corrosion probability $<10\%$ even at the initial exposure time, as presented in Figure 8(b). Conversely, within the more aggressive SCoE and SIdE electrolytes, the corrosion probability ranged between 10–90% during the initial exposure time until approximately two months, as evidenced in Figures 8(c) and 8(d). The shift from HCorZ/ICorZ to LCorZ, attained *via* the incorporation of 500 to 4000 ppm PELs into the concrete paste, enhances the corrosion resistance of the ReMS. This process facilitates the formation of a passive layer on the ReMS surface, strengthening its anti-corrosive properties. The passive film develops through physical and chemical bonding between phyto-molecules and Fe-ions, facilitated by the lone pairs of electrons in unsaturated bonds, aromatic rings, or heteroatoms of the phyto-molecules, as reported in the literature [50].

In three distinct environments – OrdE, SCoE, and SIdE – the application of PEL concentrations of 500, 1000, 2000, and 4000 ppm led to a shift in the average E_{cor} of the ReMS from the ICorZ to the LCorZ, even in all concentrations of PEL except control condition, as illustrated in Figures 9(a) to 9(d). These figures demonstrate the comparative effects of 500, 1000, 2000, and 4000 ppm PEL in three aggressive environments. The average E_{cor} in all 500–4000 ppm concentrations of PEL is higher values (*i.e.*, in a more noble direction) in OrdE, indicating better anti-corrosive action compared to the SCoE and SIdE electrolytes, based on the qualitative assessment results, as shown in Figures 9(a) through 9(d). It suggests that the effectiveness of PEL at 500–4000 ppm PEL is lower in the aggressive SCoE environment relative to OrdE and SIdE from the E_{cor} measurements.

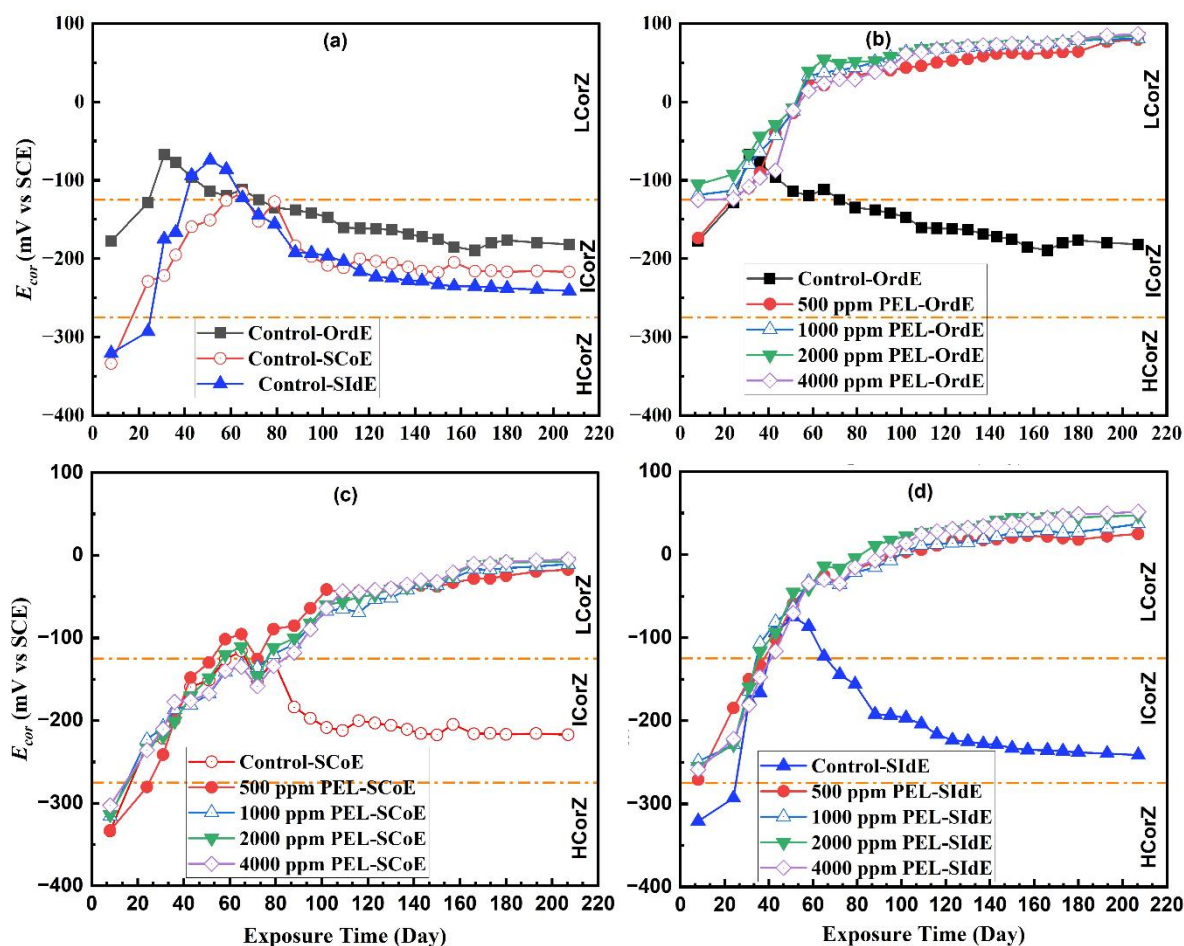


Figure 8. Changes in the corrosion potential (E_{cor}) of ReMS rod in different electrolytic solutions (a) without (control) and with 500–4000 ppm PEL at (b) OrdE, (c) SCoE and (d) SlDE conditions, as a function of exposure time.

In addition, the $EChR$ of the ReMS in RC2Bs was assessed weekly over 210 days at four surface spots per specimen, following ASTM C876-15 standard [22]. The mean $EChR$ for each RC2B was recorded in three different corrosive environments (*i.e.*, OrdE, SCoE, and SlDE) without and with 500–4000 ppm PEL, and the resultant changes in the $EChR$ are visualized, as illustrated in Figures 10(a), 10(b), 10(c) and 10(d), respectively. The findings suggest that the $EChR$ value surpassed $20 \text{ k}\Omega \cdot \text{cm}$ when 500, 1000, 2000, or 4000 ppm PEL was introduced to the RC2Bs at OrdE (Figure 10(b)), SCoE (Figure 10(c)), and SCoE (Figure 10(d)) environments after seven weeks of exposure (Figure 10(b)), predicting a region of low corrosion, consistent with the CorP results, as discussed in the previous section of this article. In contrast, the control RC2Bs without PEL in all OrdE, SCoE, and SCoE consistently demonstrated values below $5 \text{ k}\Omega \cdot \text{cm}$ (Figure 10(a)), implying a high grade of corrosiveness to ReC2Bs, in line with the E_{cor} change values depicted in Figures 8 and 9.

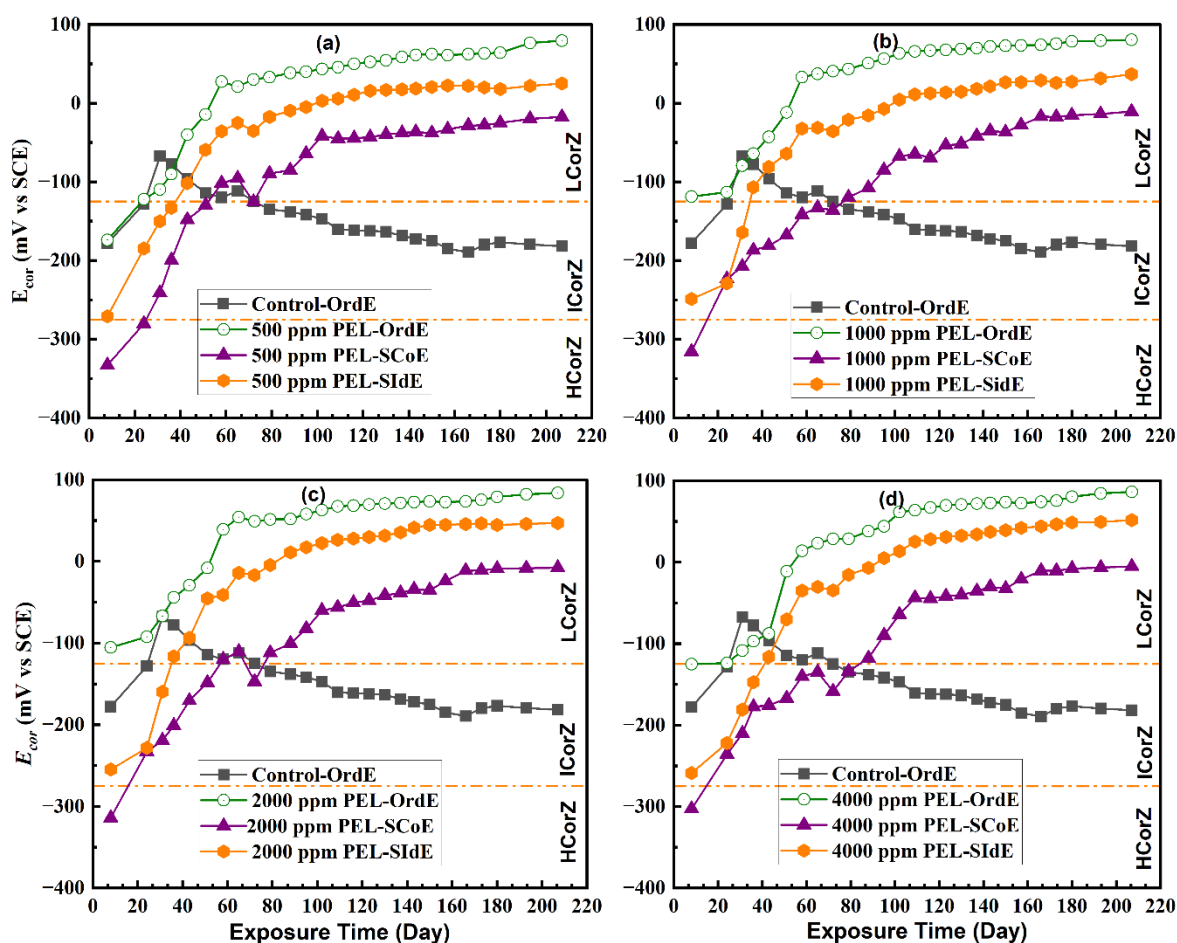


Figure 9. Variation of average corrosion potential (E_{cor}) of ReMS in concrete paste with different concentrations of PEL in ordinary (OrdE), simulated coastal (SCoE) and simulated industrial (SIde) environments, as a function of exposure time.

Figures 11(a) to 11(d) illustrated a comparative analysis of the effects of 500, 1000, 2000, and 4000 ppm PEL in three distinct environments: OrdE, SCoE, and SIde. This analysis focuses on how various concentrations influence the average electrochemical resistance ($EChR$) of the ReMS and the subsequent transition from the high corrosion zone (HCorZ) to the low corrosion zone (LCorZ). In the OrdE, the average $EChR$ across all tested concentrations of PEL exhibited significantly higher values – exceeding $20 \text{ k}\Omega \cdot \text{cm}$ – after a period of 2 to 3 months of exposure. This finding indicates superior anti-corrosive performance when compared to the SCoE and SIde environments, as illustrated in Figures 11(a) through 11(d). The data further suggest that the effectiveness of PEL within the concentration range of 500 to 4000 ppm is diminished in the more aggressive SCoE environment compared with OrdE and SIde, as indicated by the $EChR$ measurements. This effectiveness aligns with the observations derived from the E_{cor} measurements, as documented in Figures 9(a) to 9(d).

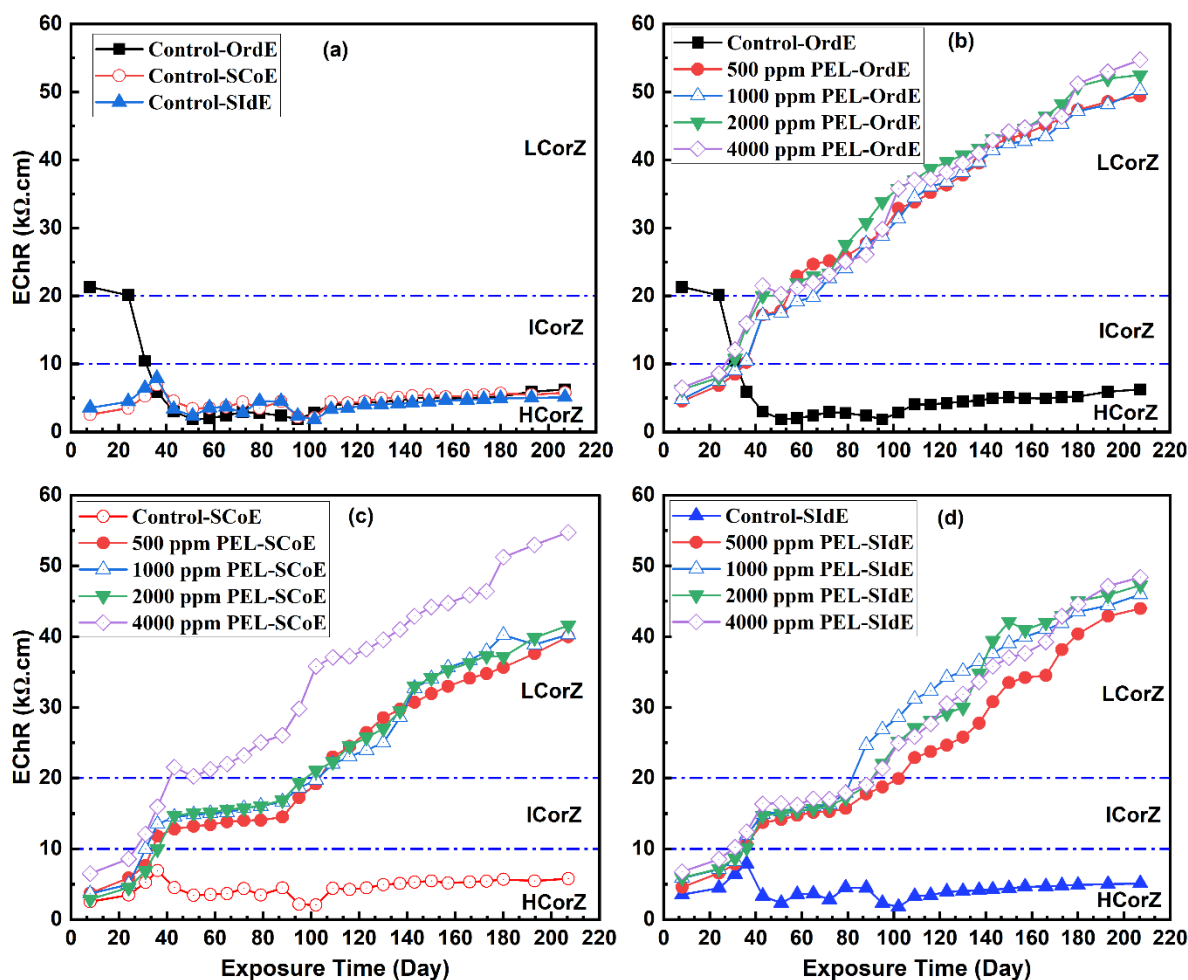


Figure 10. Variation on the average electrical resistivity ($EChR$) of ReMS at different concentrations of plant-based PEL extract (a) without and with (b) OrdE, (c) SCoE and (d) SIdE, as a function of exposure time.

The electrochemical examinations, the E_{cor} and $EChR$ measurements, exhibit that the time-varying $EChR$ approximates the foreseen results derived from the E_{cor} measurement. The $EChR$ of the RC2B samples devoid of the plant extract (control), when assessed at SCoE and SIdE conditions, is measured at less than $10 \text{ k}\Omega \cdot \text{cm}$. Additionally, the $EChR$ of the control RC2B samples, absent in the plant extract and subjected to the OrdE specially, shifts to the HCorZ after four weeks or more exposure. The speedy infiltration of corrosive ions and molecules from the outer environment *via* the concrete pore solution within four weeks of exposure is a preliminary contributor to decreasing the resistivity of the RC2Bs at OrdE conditions lacking plant extract. In the case of the control-OrdE, an ineffective film may form on the surface of ReMS after 3 weeks of exposure based on $EChR$ values, as illustrated in Figure 11. This phenomenon promotes the degradation susceptibility when subjected to aggressive ions such as Cl^- and SO_4^{2-} , which comes, respectively, from the electrolytic salts of 0.5 M NaCl and 0.5 M Na_2SO_4 .

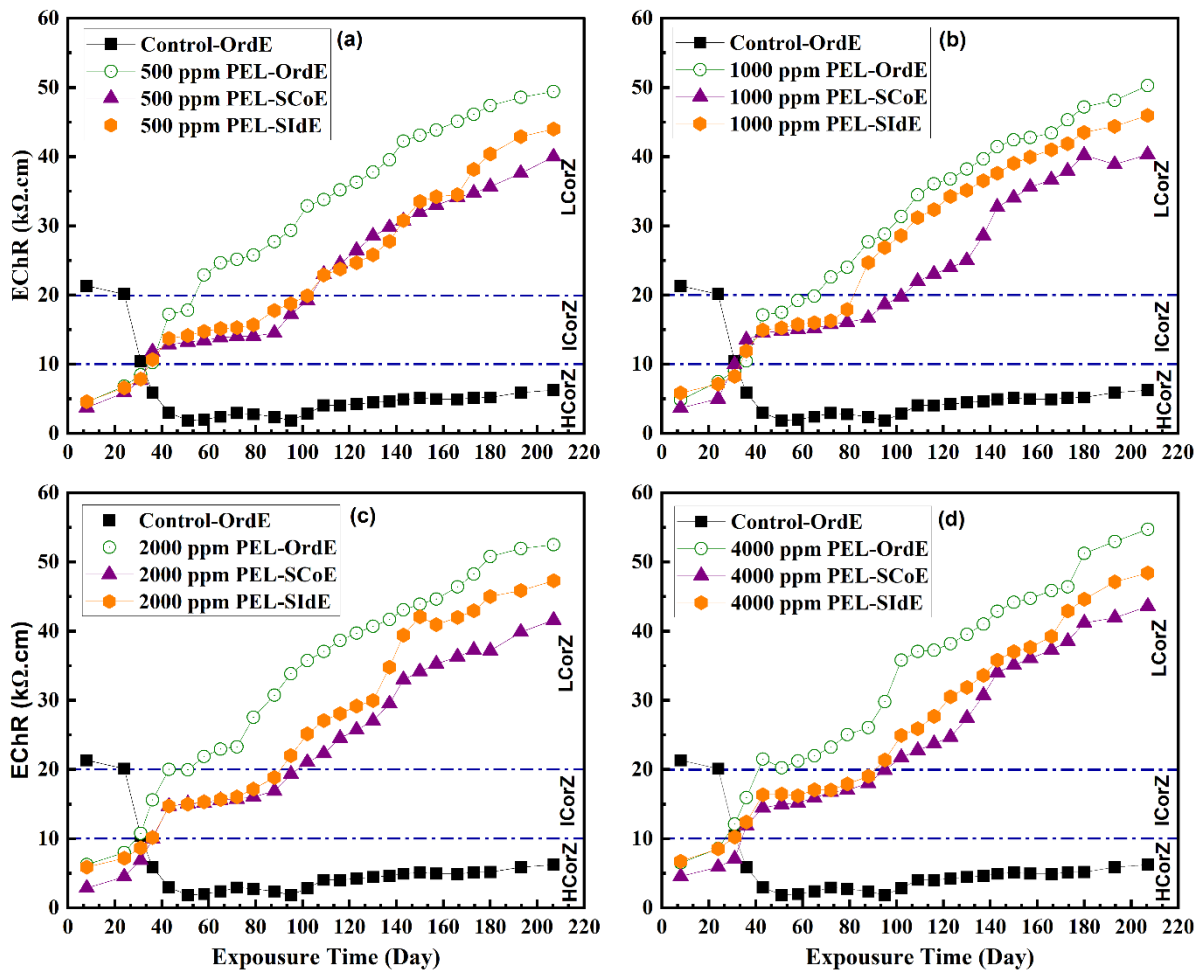


Figure 11. Variation of average electrical resistivity ($EChR$) of ReMS in concrete paste with different concentrations of PEL in ordinary (OrdE), simulated coastal (SCoE) and simulated industrial (SIde) environments, as a function of exposure time.

Therefore, the results obtained from the E_{cor} and $EChR$ measurements consistently identify three distinct levels of corrosion conditions per ASTM C876-15 standard [22]. Furthermore, the anticipated corrosion risk for the ReMS rod is evaluated based on the average $EChR$ and E_{cor} values derived from four surface points of the RC2Bs, as detailed in the existing literature [24]. High $EChR$ values indicate low conductivity, signifying ReMS passivation in concrete structures. In contrast, low $EChR$ values correspond to increased conductivity, which causes high potency of the ReCC by increasing the corrosion current. The presence of a passive film on the metal does not affect the ease of ingress of aggressive ions from the medium through concrete. However, the mixing of 500–4000 ppm PEL to concrete paste prevents the corrosion of ReMS rods by forming a barrier passive layer onto their surface, shifting average $EChR$ values to more than $20 \text{ k}\Omega \cdot \text{cm}$ in all situations, in line with the anticipated shifting of E_{cor} values to noble potential region than -126 V (SCE) , as

discussed above. At the initial four weeks, the $EChR$ values of all RC2Bs with varying amounts of PEL extracts enable them to continue within the LCorZ.

Moreover, the coexisting increase in electrochemical resistivity and corrosion potential (as reported above in Figures 8 and 9) could indicate a direct correlation with the densification of concrete in the presence of phyto-molecules [51]. The densification process reduces water content and enhances the packing of concrete, with a decreased porosity; the number of pores decreases, leading to higher resistivity, as reported in previous work [52]. The observed increases in the $EChR$ and E_{cor} values of RC2Bs with exposure time, following the addition of PEL at the conditions of OrdE, SIdE, or SCoE, signify a substantial enhancement in their $EChR$ and E_{cor} values over time. Such effects have the potential to diminish the permeation of corrosive ions into the surface of ReMS rods in the presence of 500–4000 ppm PEL, even in various aggressive environments. The practical implications of these findings suggest a promising opportunity to improve the longevity and durability of ReCIs.

3.5. Analysis of OM, SEM-EDS and 3-D WLI images

Except for the electrochemical investigations, optical microscopy (OM) and scanning electron microscopy (SEM) were employed to examine the surface morphology of corroded ReMS after treatment with different severe environments of the concrete composite, providing illustrative evidence of the corrosion inhibition mechanism, as explained elsewhere [53]. Figure 12(a) displays the optical microscopy (OM) image of a fresh MilS rod before reinforcement in RC2B. In contrast, Figures 12(b), 12(c), and 12(d) show OM images of the MilS after being reinforced in concrete without PEL extract and exposed to ordinary (OrdE), simulated industrial (SIdE), and simulated coastal (SCoE) environments for approximately seven months. Analysis reveals a higher presence of corrosion products on the ReMS surfaces subjected to industrial and coastal environments compared to those in ordinary conditions. However, after the addition of 4000 ppm PEL to the concrete mix at OrdE, SIdE, and SCoE environments for seven months, the ReMS did not exhibit any significant corrosion spots, as illustrated in Figures 12(e), 12(f), and 12(g), respectively. These OM images were of 20 times magnification and do not reflect compositional variations. Therefore, we conducted further surface analysis using SEM-EDS methods.

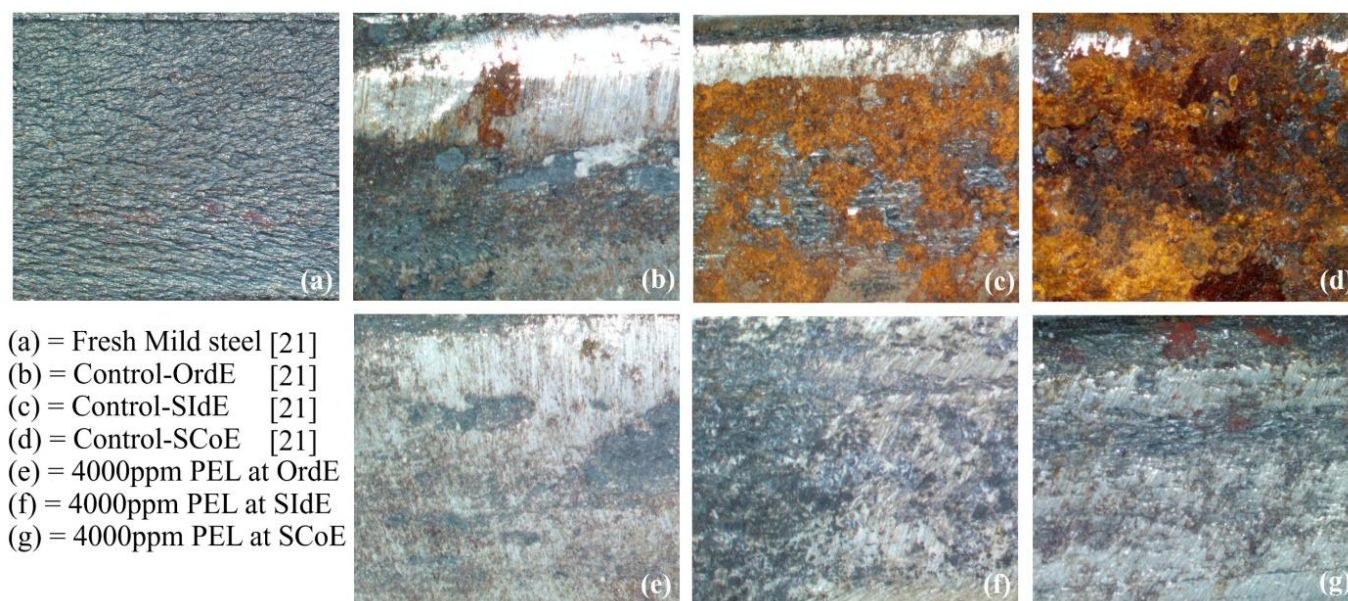


Figure 12. Optical microscopic images of surface layers formed on a fresh MilS (a); ReMS at control-OrdE (b), control-SIdE (c), and control-SCoE (d), ReMS in RC2Bs with 4000 ppm PEL at OrdE (e), with 4000 ppm PEL at SIdE (f) and with 4000 ppm BEL at SCoE (g) after seven months exposure.

The SEM coupled with energy-dispersive X-ray spectroscopy (EDS) analyses has substantiated the presence of corrosion protective layers on the surface of ReMS in RC2Bs. Therefore, the SEM examination technique investigated further the morphological changes of the ReMS rod surface after it was embedded for about seven months in an RC2B with and without 4000 ppm PEL in various setting electrolytes like OrdE, SCoE, and SIdE, including the fresh MilS rod surface analysis. A comparatively smooth surface was found in the fresh MilS rod, as shown in Figure 13(a). Also, as seen in Figures 13(b) and 13(c), the SEM images of the control surface in two unlike aggressive surroundings (*i.e.*, OrdE and SCoE, respectively) reveal patches of rust covering large portions of the ReMS surfaces after being exposed for about seven months. However, as illustrated in Figures 13(d), 13(e), and 13(f), the rust-induced irregular and flawed surfaces are characterized by a relatively minimal to moderate degree of smoothness on the ReMS rods. No significant corrosion damage to the ReMS surface is anticipated in the concrete pore solution with a concentration of 4000 ppm PEL, in contrast to the SEM images of the reinforcing metal observed in control-RC2B, which did not incorporate the plant extract.

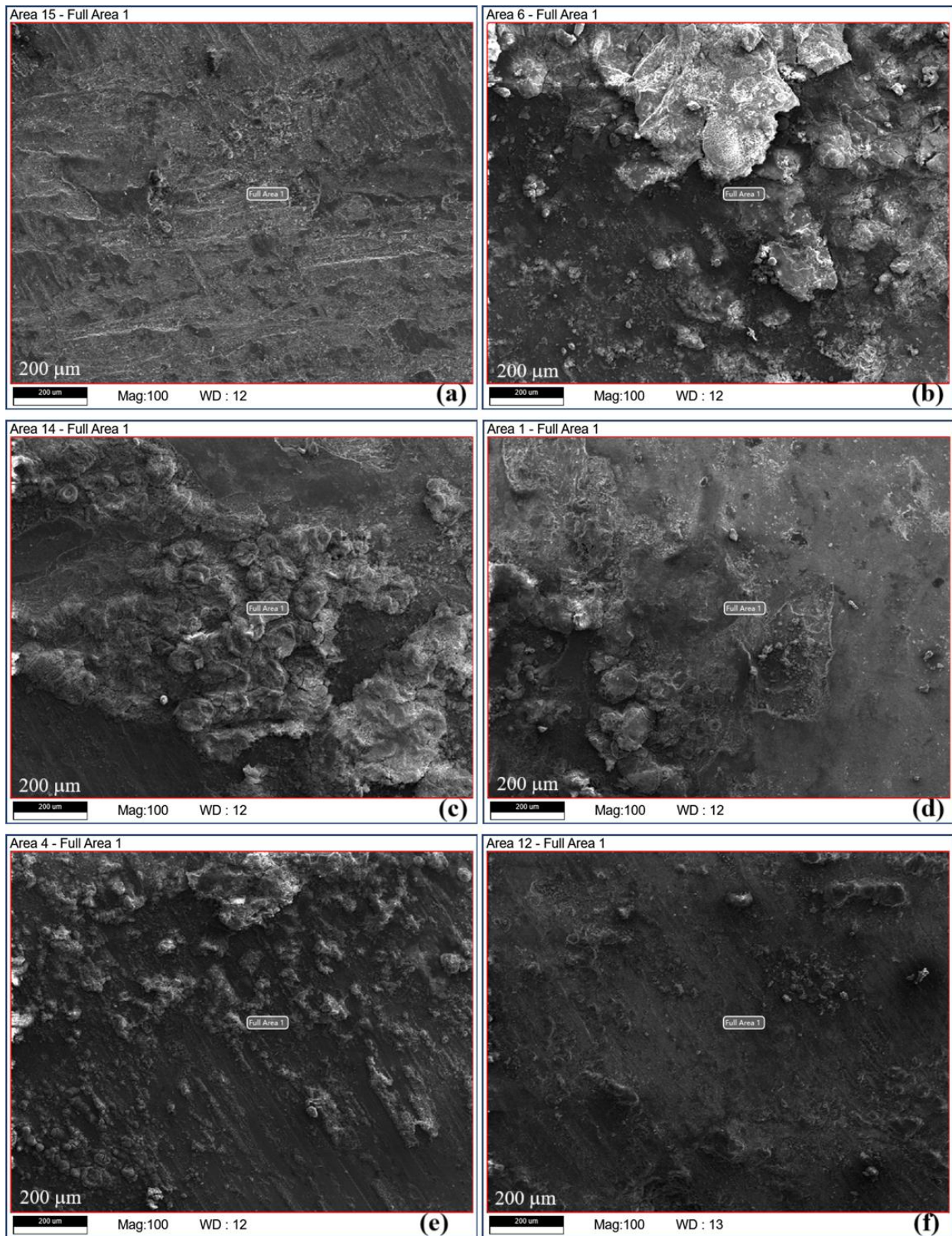


Figure 13. SEM images of the fresh MS rod surfaces (a), ReMS in control OrdE (b), ReMS in control SCoE (c) ReMS in RC2B with 4000 ppm PEL at SCoE (d), ReMS in control SIdE (e), and (f) ReMS in RC2B with 4000 ppm PEL at SIdE (f) after being exposed for seven months.

According to earlier research [54], the phyto-molecules of the plant extracts act as an adsorbent to form a protective passive film on the active corrosion sites of the ReMS surface to leave smooth, continuous layers that protect it against corrosion. The SEM images definitively illustrate that the surface imperfections associated with corrosion have been eliminated on the inhibited surface. This observation confirms the superior inhibition effectiveness of the PEL extracts in corrosive environments, particularly those that contain chloride and sulfate in concrete. Consequently, the probability of corrosion damage to the ReMS has transitioned from the active or HCorZ to a passive or LCorZ following the incorporation of 500–4000 ppm PEL into the concrete paste. This finding is supported by prior reports concerning the bark extract from the same plant [21]. Besides, these SEM results are consistent with the electrochemical data previously discussed.

Furthermore, the elemental analysis conducted using SEM-EDS reveals that the weight percentage of iron (Fe wt.%) in the ReMS material lowers following its embedment in RC2Bs. This lowering is particularly evident when 4000 ppm of PEL is utilized as a concrete admixture within the OrdE, SCoE, or SIdE environments, accompanied by increased oxygen coverage. This observation corroborates the findings from the compositional variations identified in the EDS study. Figures 14(a), 14(b), and 14(c) present the EDS analysis, along with compositional interpretations, for the surface of the fresh MilS rod before the implantation of the ReMS rod, as well as after a period of 7 months of implantation in RC2Bs, at controlled conditions for both OrdE and SCoE environments, incorporating 4000 ppm of PEL. Uniformly severe corrosion and the production of adhering corrosion products formed on the ReMS surface after exposure for seven months in corrosive control OrdE and SCoE conditions without the PEL extract, as illustrated in Figures 14(b) and 14(c), respectively. It happens by a continuous oxidation loss of the ReMS.

However, Figures 14(d), 14(e), and 14(f) represent the EDS spectra analysis for the ReMS rod after 7 months of embedding in concrete beams with 4000 ppm PEL in control-OrdE, control-SCoE, and control-SIdE, respectively. The composition of MilS rods, before and during their embedding in concrete, is compared to that of ReMS rods in RC2Bs exposed to varying corrosive environments. The surface of corroded ReMS rods exhibits markedly lower iron (Fe) amounts when compared with fresh MilS composition. Conversely, there is a significant presence of oxygen (O) atoms in all ReMSs within the RC2Bs at the control conditions of OrdE, SCoE, and SIdE in the absence of PEL extract. This observation indicates the formation of iron oxide as a corrosion product [55]. Based on these findings, it can be speculated that ReMS rods exposed to the control-SCoE demonstrate a higher susceptibility to corrosion than those evaluated at the control-SIdE, while the control-OrdE condition is suspected to be less corrosive in nature.

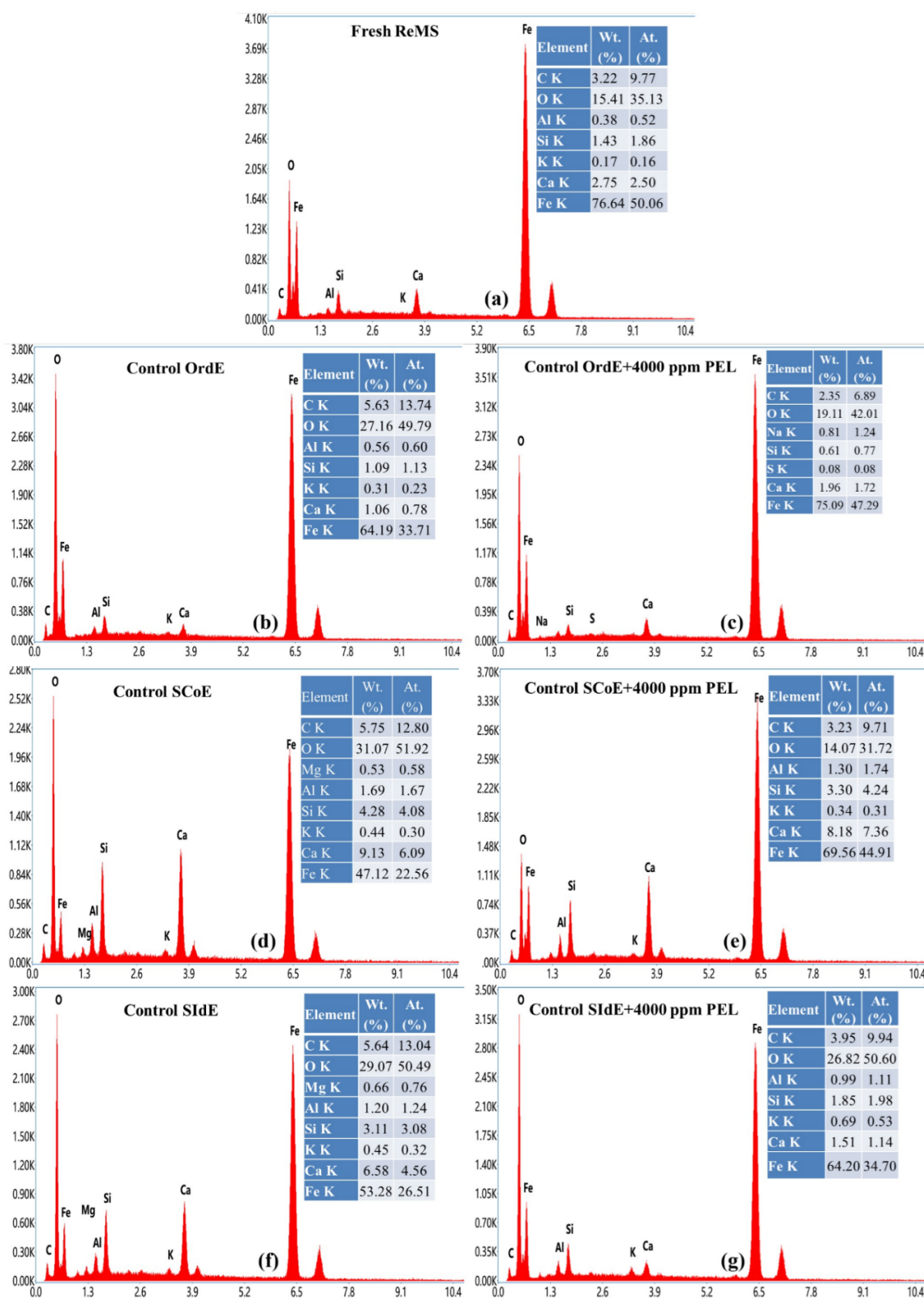


Figure 14. EDS spectra analysis of (a) fresh MILS rod surface before embedding and ReMS rod after embedding for 7 months in RC2Bs at (b) control-OrdE, (c) OrdE with 4000 ppm PEL, (d) control-SCoE, (e) SCoE with 4000 ppm PEL, (f) control-SIdE, and (g) SIdE with 4000 ppm PEL.

In summary, the SEM-EDS results are agreeing with those outcomes from the E_{cor} and E_{ChR} measurements, proving that PEL extract functions effective corrosion inhibitor. The PEL extract holds potential concrete admixture for corrosion control application within marine and industrial sectors, including normal conditions, and may serve as a viable alternative to other widely utilized corrosion inhibitors. Nevertheless, more research is crucial to evaluate the long-term stability of the PEL-Fe complex film, its efficacy at varying environmental conditions, and its compatibility with other corrosion prevention strategies. Such investigations are crucial for realizing the full potential of PEL extract as a corrosion protection solution for the ReCC.

Besides the SEM/EDS examination, the three-dimensional (3D) topography approach of white light interferometry (WLI) can be explored for investigating 3D WLI images of the tested ReMS specimens, prepared after exposing the RC2Bs for 210 days in three different environments without (*i.e.*, control-OrdE, control-SCoE, and control-SIdE) and with 4000 ppm PEL in concrete blends at OrdE, SCoE, and SIdE as concrete admixture, as illustrated in Figure 15. WLI offers rapid, meticulous, non-contact 3D surface topography critique for characterizing materials. It provides exceptional lateral and vertical resolution [56], making it a valuable tool for precision surface analysis. WLI can outline the surface profile of the ReMS structures, even on smooth surfaces with minimal roughness measured on the nano-scale.

The 3D topographical WLI images of the ReMS provide valuable information about the surface roughness of the reinforcing metal after exposure for seven months in distinct control environments without and with 4000 ppm PEL. The average roughness (S_a) of the test ReMS specimens implanted in three control corrosive circumstances reported in a range of 9.385–5.856 μm (Figures 15(a), 15(c), 15(e)), which have significantly high values compared with the S_a value of the ReMS specimens treated with 4000 ppm PEL at the corresponding environment (Figures 15(b), 15(d), 15(f)), presumably due to severe corrosion in these three control conditions. The S_a values for the ReMS samples at the control-OrdE without and with 4000 ppm PEL are recorded at 6.311 μm and 2.799 μm , respectively, as illustrated in Figures 15(a) and 15(b). Similarly, the S_a values for the ReMS samples at the control-SIdE, without PEL and with 4000 ppm PEL, are 9.385 μm and 3.021 μm , respectively, as depicted in Figures 15(c) and 15(d), while the S_a values for the ReMS specimens subjected to 4000 ppm PEL at SCoE without and with 4000 ppm PEL are 5.856 μm and 3.449 μm , respectively, as depicted in Figures 15(e) and 15(f), respectively, demonstrating reductions of approximately 67.81% at SIdE and 69.79% compared to the values observed in the respective control-SCP solutions. This finding suggests that PEL exhibits superior corrosion-inhibiting properties and effectively mitigates the early degradation of reinforcing mild steel in RC2Bs.

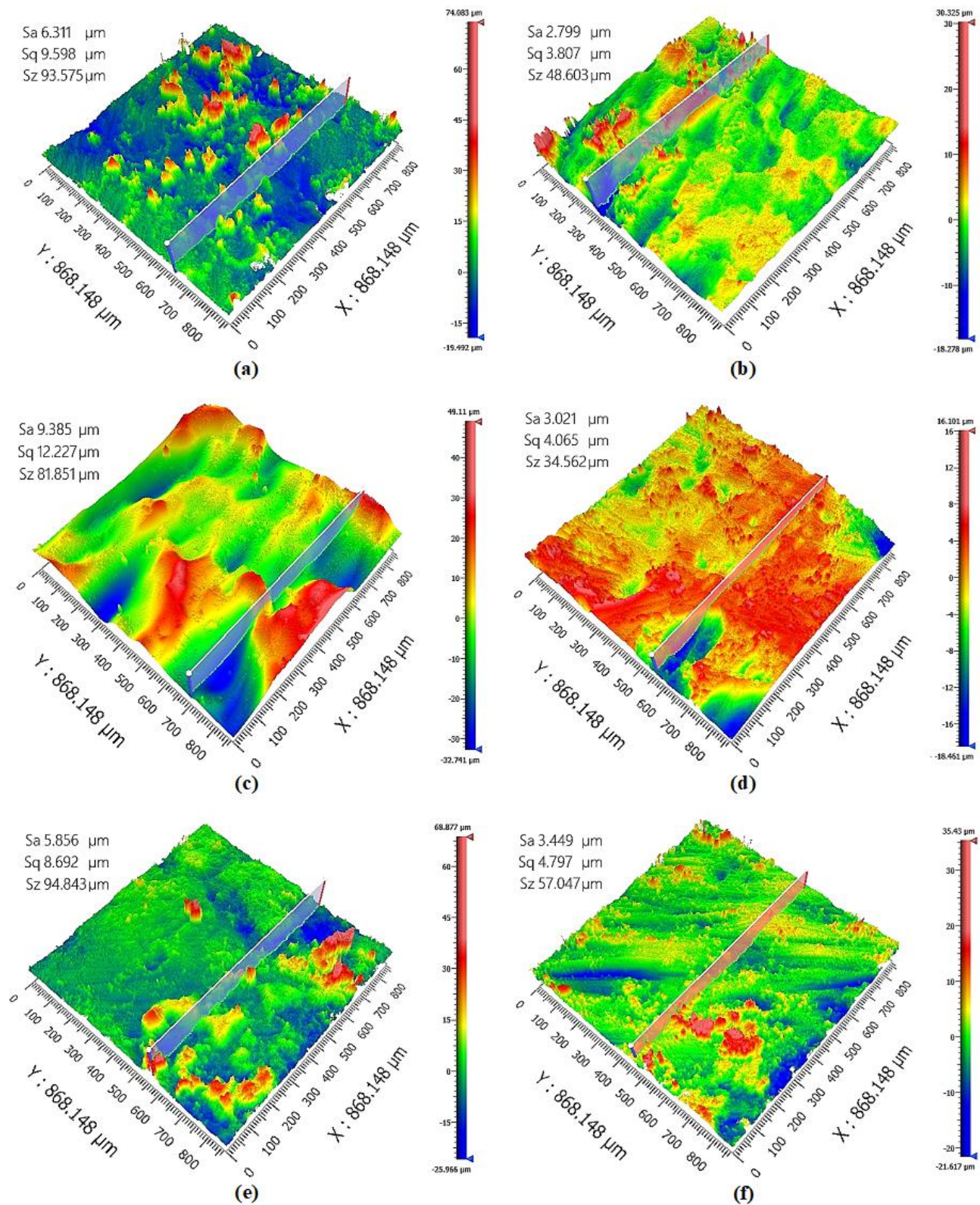


Figure 15. Three dimensional (3-D) WLI images of ReMS surfaces after 7 months embedded in RC2Bs at control-OrdE (a), 4000 ppm PEL at OrdE (b), control-SIdE (c), 4000 ppm PEL at SIdE (d), control-SCoE (e), and 4000 ppm PEL at SCoE (f).

3.6. Corrosion inhibition mechanism

A visual representation illustrates a plausible corrosion-inhibiting mechanism of ReMS at concentrations ranging from 500 to 4000 ppm PEL at three environmental conditions: OrdE, SCoE, and SIde, as illustrated in Figure 16. This convincing model is proposed based on a comprehensive array of experimental procedures, including screening tests, UV-Vis, FTIR, GC-/UHPLC-MS analyses, E_{cor} , E_{ChR} , and various 2-D and 3-D surface analysis techniques, such as OM, SEM/EDS, and WLI. The morphology of the ReMS surface becomes smoother as the concentration of PEL in the concrete paste increases, which reduces the number of defect points in the protective layers. The corrosion-preventive passive film is primarily attributed to the effective adsorption of the inhibitor molecules, which decreases the concentration of corrosive ions such as Cl^- or SO_4^{2-} on the ReMS surfaces. They are replaced by electron-rich polar or functional groups, including $-\text{OH}$ groups in aromatic rings, $-\text{NH}_2$, $-\text{CH}_2\text{OH}$, cyclic ethers, and amides. These functional groups enhance adsorption onto the ReMS surface. The plausible corrosion-inhibiting mechanism, complemented by such adsorption phenomena, arises from the displacement of aggressive ions and molecules from the ReMS surface, as illustrated in Figure 16. The interaction facilitated through the sharing of electrons between the electronegative oxygen, nitrogen, and sulfur components of the PEL extract and the iron substrate – *via* chemical, physical, or a combination of both adsorption types – contributes to the enhanced protection of ReMS against ReCC.

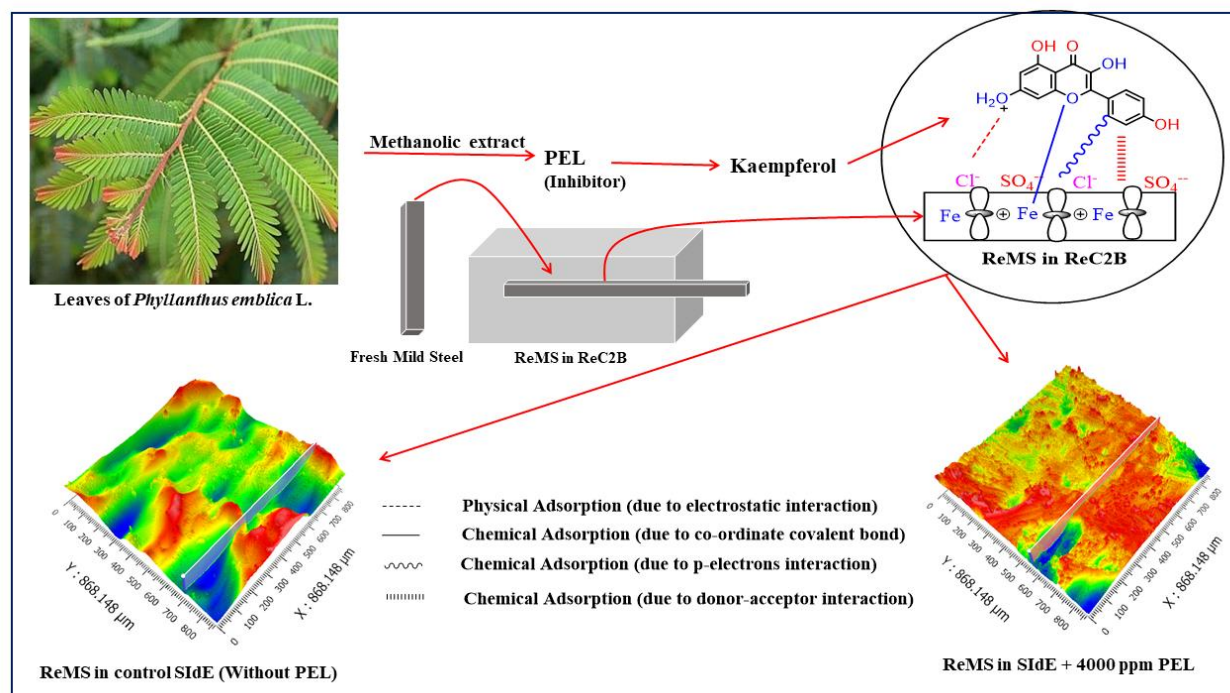


Figure 16. Schematic graph showing corrosion inhibiting mechanism by phytochemicals present in PEL.

Chemisorption occurs when unshared electron pairs from heteroatoms, such as oxygen, nitrogen, and sulfur present in phyto-molecules, form coordinate covalent bonds [57]. It facilitates interactions between the π -electrons of aromatic rings in phyto-molecules and the vacant d-orbitals of iron (Fe) atoms on the ReMS rod. In contrast, physisorption, characterized as a form of electrostatic adsorption occurs when charge-carrying phyto-molecules – such as phenols, alkaloids, flavonoids, and terpenoids – interact with Fe^{2+} ions on the surface of the ReMS through electrostatic bonding [58]. As a result, various phyto-molecules form a uniform and stable corrosion-protective passive film between the corrosive concrete composite and the ReMS surfaces. This study explores different phyto-molecules extracted from the leaves of the indigenous plant *Phyllanthus emblica* (L.), native to Nepal, to formulate concrete admixtures.

4. Conclusions

Plant-based, eco-friendly admixtures for concrete corrosion have garnered significant interest in recent years as a viable solution to the challenges associated with the premature failure of various ReCIs. The study explored the outcome of a leaf-based PEL concrete admixture for controlling reinforced concrete corrosion (ReCC) through the techniques of electrochemical corrosion potential and resistivity measurements, and surface analysis using OM, SEM/EDX and WLI. The electrochemical analyses of corrosion potential and electrical resistivity measurements validated the efficacy of the PEL extract as a concrete admixture based on the surface-sensitive two-dimensional SEM/EDS and three-dimensional WLI analyses.

The methanolic extract formulated from the leaf powder of the *Phyllanthus emblica* (L.) plant (PEL) exhibits meritorious properties for controlling concrete corrosion of ReMS at three environments: OrdE, SCoE, and SIdE. Satisfactory anti-corrosive efficacy is reported between 500 and 4000 ppm PEL, even though the PEL functions as concrete admixture at 500–2000 ppm. Chemical screening tests and spectroscopic techniques ensure PEL retains alkaloids, flavonoids, phenols, and steroids as main phyto-compounds in SMBs.

Electrochemical measurements indicate a trend where the E_{cor} and E_{ChR} shift positively, demonstrating noble behavior. The extract from *Phyllanthus emblica* (L.) leaves (PEL) might be a sustainable and effective corrosion inhibitor, capable of mitigating early corrosion damage in ReCIs. The findings of this study may lead to the development of plant-based anti-corrosive concrete admixtures on a commercial scale, potentially replacing synthetic waterproofing chemicals. Future research should focus on a more detailed investigation of the corrosion inhibition mechanism using techniques such as electrochemical impedance spectroscopy (EIS), linear polarization, and other relevant surface analysis methods.

Acknowledgements

The authors acknowledge the UGC-Nepal for providing MG's PhD assistance support (PhD-78/79-S&T-03). The University of Lucknow-India (Mr. Anant Ram Rathor and Mr. Gulab Singh), University of Cincinnati (Dr. M.A. Fickenscher), Cincinnati, OH-USA, and IMR-CAS, Shenyang of China are thankful for providing supportive analytical facilities; XRD & UHPLC-MS, SEM/EDS, and WLI, respectively.

Funding

Also, the work partially aided the JB-NPB-KPB team of the Central Department of Chemistry-TU through the Ministry of Education, Science and Technology (MoEST), Government of Nepal, Singh Darbar, Kathmandu under the "Research and Technology Grants (Material Science)" for the fiscal year 2080/081 (2023/024 AD).

CRedit authorship contribution statement

MG: conceptualization, experiment design, sample preparation, investigation, data analysis, draft writing; JRD: sample preparation, instrumental analysis; DBS: data analysis, validation; MD: surface analysis, interpretation; KPB: data analysis, validation, draft writing; NPB: conceptualization, validation, results summarization, draft writing, supervision; JB: review & final manuscript – writing, conceptualization, validation, supervision.

Conflicts of interest

The authors claim that they have no competing interests.

Ethical statement

This study's experiment did not involve human subjects, so there was no need to obtain ethical clearance from our institutional ethics board.

Data availability statement

Upon request, the corresponding authors will provide the supporting data.

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