

Enhanced anti-biocorrosion performance of a superhydrophobic Ni-P/PDMS-SiO₂ organic-inorganic hybrid composite coating against *Pseudomonas aeruginosa*

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Abstract: Microbiologically influenced corrosion (MIC) is a major challenge for extending the service life of X65 pipeline steel, especially in environments with high concentrations of *Pseudomonas aeruginosa*. This study prepared a Ni-P/PDMS-SiO₂ hierarchical composite coating to extend protection by creating a dual-barrier defense system. Systematic electrochemical tests over 45 days showed that both bare X65 steel and the single Ni-P intermediate layer deteriorated rapidly. However, the composite coating maintained a relatively high level of low-frequency impedance modulus ($|Z|_{0.01}$ Hz), approximately $10^{10} \Omega \cdot \text{cm}^2$. It was six orders of magnitude greater than that of bare steel and did not show any significant change after being immersed in the *P. aeruginosa*-containing medium. Based on these results, it could be assumed that a stable Cassie-Baxter air plastron and a dense amorphous Ni-P interlayer have a combined effect of physically blocking bacterial attachment and metabolite infiltration, thereby providing a support structure. This paper offers a feasible approach to designing extended-life anti-MIC coatings in bacterial-laden environments.

Key words: Superhydrophobic surface; Organic-inorganic hybrid coating; *Pseudomonas aeruginosa*; Microbiologically influenced corrosion; Pipeline protection

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1. Introduction

Due to its excellent mechanical strength and cost-effectiveness, the oil and gas extraction industry highly relies on X65 pipeline steel [1]. However, the actually used pipelines are often affected by microbiologically influenced corrosion (MIC), which is a common degradation mechanism. Its degree of causing or accelerating local structural failure is much higher than non-biological corrosion [2-4]. *Pseudomonas* species, especially *Pseudomonas aeruginosa* [5], is extremely common in industrial and marine environments and plays a pioneering colonization role in the early stage of biofilm formation [6, 7]. This microorganism consumes oxygen and accumulates corrosive metabolic products, creating a harsh local microenvironment that ultimately destroys the protective passive film and accelerates the dissolution of the base metal [8-10]. Therefore, effective control of MIC is crucial for maintaining pipeline integrity.

To prevent the pipeline infrastructure from being affected by MIC, functional surface coatings are widely regarded as an effective protective measure [11, 12]. However, the traditional single-layer protection strategy has significant flaws. Organic

polymer coatings (such as polyurethane and epoxy resin) can provide adequate physical shielding effects, but their surface hardness is relatively low. Under external mechanical stress, these coatings are prone to develop microcracks, creating local gaps that provide channels for bacterial colonization [13]. Conversely, the electroless high-phosphorus (with a phosphorus content exceeding 9 wt%) nickel-phosphorus (Ni-P) coatings are renowned for their high hardness and excellent chemical protection properties, and are particularly suitable for harsh environments [14-17]. Although these pure inorganic layers have a dense amorphous structure, long-term use inevitably leads to microdefects, thereby forming new pathways for electrolyte penetration and bacterial adhesion [18]. Therefore, the traditional single-layer structure cannot meet the long-term requirements for MIC environments.

Recently, biomimetic highly hydrophobic materials have been introduced to solve the MIC problem of metal [19, 20]. These materials simultaneously achieve micro/nano-scale layered rough structures and low surface energy modification, thereby forming a stable non-wetting surface that physically isolates the metal from corrosive fluids and bacterial secretions. Although many coatings have shown excellent initial anti-corrosion and anti-bacterial properties in the laboratory [21-23], they often have poor mechanical properties and are difficult to withstand harsh conditions. The fine micro/nano structures are highly susceptible to industrial shear, wear, and impact stress, leading to rapid deterioration of the anti-corrosion interface and inability to provide protection. Therefore, improving

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the mechanical stability of these coatings remains a key research challenge.

Therefore, a gradual organic-inorganic hybrid system that combines the properties of both rigid and flexible materials has been proposed. In this paper, a composite coating was designed to provide combined protection for X65 steel against *P. aeruginosa*. It is different from the previous superhydrophobic anti-corrosion coatings. Traditional superhydrophobic systems are usually of an overall structure or pure polymer structure, relying solely on surface defense mechanisms. Due to physical wear and tear, the micro-wetting interface is damaged, and the base material will rapidly corrode. In contrast, this system introduces a hard, amorphous Ni-P inorganic intermediate layer and a low surface energy PDMS-SiO₂ surface layer. At the same time, a stable multilayer surface structure is constructed using nano-sized SiO₂ [24–26], effectively inhibiting bacterial adhesion and preventing the initial formation of biofilms.

2. Materials and Methods

2.1. Substrate pretreatment

X65 pipeline steel was processed into a test coupon of appropriate size (10 mm × 10 mm × 5 mm). The surface was pretreated sequentially to ensure strong coating adhesion. First, the X65 coupons used 600, 800, 1000, 1200 and 2000 SiC abrasive paper for wet grinding. Then rinsed X65 coupons with deionized water and absolute ethanol before being dried with a cold nitrogen stream to prevent oxidation. Organic pollutants and abrasive debris were removed by ultrasonic treatment of X65 coupons in absolute ethanol for 5 min. Subsequently, the steel substrates were degreased in a solution containing 3 wt% NaOH and 3 wt% Na₂CO₃ at 60°C for 20 min. Finally, the X65 coupons were immersed in a 10 wt% HCl solution for 50 s for surface activation, rinsed with deionized water, and stored for use.

2.2. Coating fabrication procedure

Reagents employed in the electroless Ni-P bath included citric acid (C₆H₈O₇), succinic acid (C₄H₆O₄), sodium hypophosphite monohydrate (NaH₂PO₄·H₂O), lactic acid (C₃H₆O₃), nickel sulfate hexahydrate (NiSO₄·6H₂O), sodium saccharin dihydrate (C₇H₄NNaO₃S·2H₂O), sodium dodecyl sulfate (C₁₂H₂₅SO₄Na), lead nitrate (Pb(NO₃)₂), and NaOH for pH adjustment. To form the Ni-P interlayer, the pretreated substrate was immersed in the bath containing 20 g/L C₆H₈O₇, 14 g/L C₄H₆O₄, 30 g/L NaH₂PO₄·H₂O, 20 mL/L C₃H₆O₃, 25 g/L NiSO₄·6H₂O, 0.06 g/L C₇H₄NNaO₃S·2H₂O, 0.03 g/L C₁₂H₂₅SO₄Na, and 1 mg/L Pb(NO₃)₂. The pH was adjusted to 6.0 using NaOH. The deposition process was maintained at (85 ± 1)°C for 2 h under continuous mechanical agitation at 200 rpm. Finally, the samples were sonicated in absolute ethanol for 5 min to eliminate the solution residues and then dried [27].

The preparation process of the composite coating was improved based on our previous research [28]. N-hexane was used to reduce the high viscosity of the PDMS precursor for the PDMS-SiO₂ composite layer at a 1:1 (v/v) ratio. Superhydrophobic SiO₂ nanoparticles were added at concentrations of 0.1–5 wt%. Taking the 1 wt% formulation as an example, 0.1 g of superhydrophobic SiO₂ was dispersed in 10 mL of n-hexane via ultrasonic agitation, and this suspension was combined

with 10 mL of PDMS prepolymer and 1 mL of curing agent. The mixture was homogenized using an ultrasonic probe to break up agglomerates. The resulting slurry was then coated on the Ni-P layer. A vacuum was used to draw the mixture into the interstices and pores. After solvent evaporation, the samples were thermally cured in an oven at 80°C for 2.5–3 h to obtain the composite coating (as shown in Fig. 1).

2.3. Structural and surface characterization

The morphology and elemental distribution of the coating surface and cross-section were examined using Scanning Electron Microscopy (SEM; Sigma 560, Zeiss, Germany) equipped with Energy Dispersive X-ray Spectroscopy (EDS). Sputter-coat samples that have non-conductive polymeric surfaces with a thin gold layer before imaging to prevent charging. X-ray Diffraction (XRD; Ultima IV, Rigaku, Japan) in a Cu K α radiation environment ($\lambda = 0.15406$ nm) was carried out at 10° to 80° to determine the crystal structure and phase stability of the Ni-P interlayer. Fourier Transform Infrared Spectroscopy (FTIR; Nicolet iS50, Thermo Scientific, USA) was used in ATR mode from 4000 cm⁻¹ to 400 cm⁻¹ to identify the characteristic functional groups (such as Si-O-Si and -CH₃) of the hybrid layer.

Surface wettability was quantified at ambient temperature using an optical contact angle goniometer (Model 250, ramé-hart, USA). A 5 μ L droplet of deionized water was used as the probe liquid.

2.4. Anti-microbial corrosion assessment

2.4.1. Bacterium and culture medium

Anti-microbial corrosion resistance of the composite coatings was evaluated using the *Pseudomonas aeruginosa* strain. The bacteria were cultured in an LB-NO₃ medium, composed of 10 g tryptone, 10 g KNO₃, 5 g yeast extract, and 5 g NaCl per 1 L of DI water [4]. The initial pH of medium was adjusted to 7.2 ± 0.1 using NaOH. The culture medium, test vials, rubber stoppers, and required experimental tools were sterilized via autoclaving at 121°C for 30 min prior to inoculation. Furthermore, the L-cysteine solution was filter-sterilized using a 0.2 μ m Stericup membrane (Thermo Scientific Nalgene, USA) and added into the medium.

2.4.2. Biofilm viability observation

At 37°C, pre-incubate the *P. aeruginosa* strain in an incubator for 3 days [29]. The initial density of the planktonic cell was about 10⁶ cells/mL. The coated test samples were then added to the active *P. aeruginosa* medium mentioned above, and after incubation at 37°C, the initial anti-biofouling effect was observed.

To evaluate bacterial adhesion and biofilm formation, the exposed samples were gently rinsed with sterile phosphate-buffered saline (PBS) to remove the planktonic cells. Subsequently, the surface sessile cells were stained using a LIVE/DEAD® BacLight Bacterial Viability Kit (Invitrogen, Eugene, OR, USA) for 15 min in the dark. The viability of sessile cells was then characterized using confocal laser scanning microscopy (CLSM; A1RHD25, Nikon, Japan). Finally, quantitative analysis of the acquired fluorescence images was executed using ImageJ software.

2.4.3. Electrochemical measurements

Electrochemical measurements of the samples in *P. aerugi-*

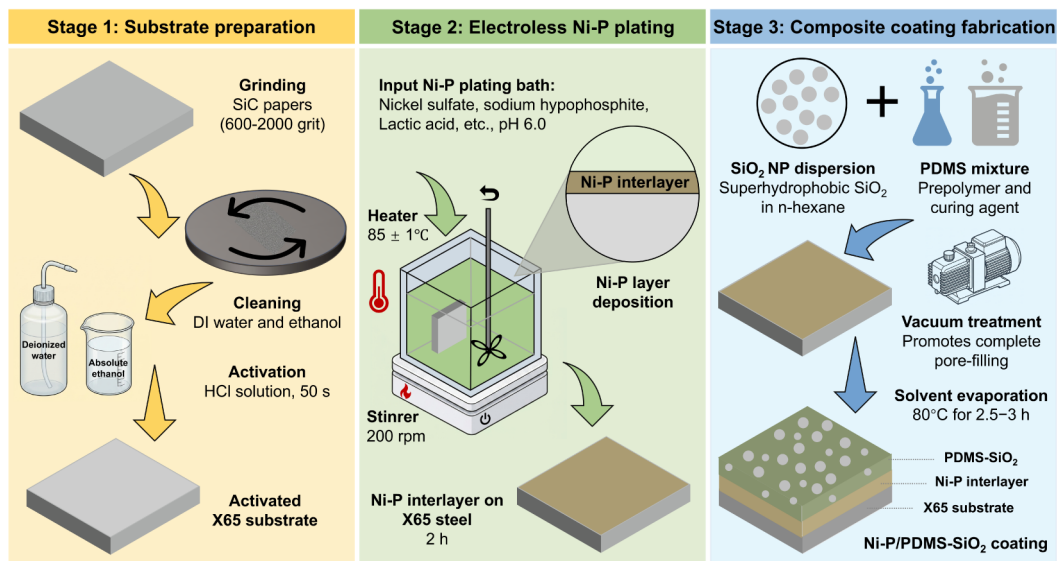


Fig. 1. Preparation process and structural schematic diagram of the hierarchical superhydrophobic Ni-P/PDMS-SiO₂ composite coating on X65 steel substrate.

nosa-containing medium were conducted at a Gamry Reference 600+ workstation (Gamry Instruments, Philadelphia, PA, USA). A typical three-electrode system was used [30], with the test sample as the working electrode (1 cm² exposed area), a saturated calomel electrode (SCE) as the reference, and a platinum plate (2 cm × 2 cm) as the counter electrode.

The electrochemical testing sequence was sequentially executed as follows. "First, the open circuit potential (OCP) was continuously recorded for more than 30 min to ensure that a quasi-steady-state thermodynamic condition was achieved. Stabilization was carried out, and then at the OCP, electrochemical impedance spectroscopy (EIS) was conducted in the frequency range of 10⁵ Hz to 10⁻² Hz with an AC sinusoidal perturbation of 10 mV. Suitable equivalent circuit models (ECMs) were selected to fit the obtained EIS data and extract kinetic parameters. Electrochemical tests were conducted in triplicate, and multiple sets of data were obtained for statistical anal-

ysis.

3. Results and Discussion

SEM and EDS were used to examine the surface morphology and element distribution of the prepared coatings. Figs. 2a and 2b show the surface features of the electroless Ni-P interlayer. A typical nodular "cauliflower-like" structure was the interlayer, and these nodules were evenly and densely distributed. This is the structure of a well-deposited amorphous Ni-P coating. As shown in the corresponding EDS map, the Ni and P elements were evenly distributed at the surface, and there were no prominent impurities (Figs. S1a–S1a"). The morphological transition after the application of PDMS-based topcoats is shown in Figs. 2c and 2d. The pure PDMS coating (Fig. 2c) was relatively flat and even in this aspect. Hydrophobic SiO₂ nanoparticles were used to modify the surface structure to some extent. As shown in Fig. 2e, the SiO₂ particles formed clus-

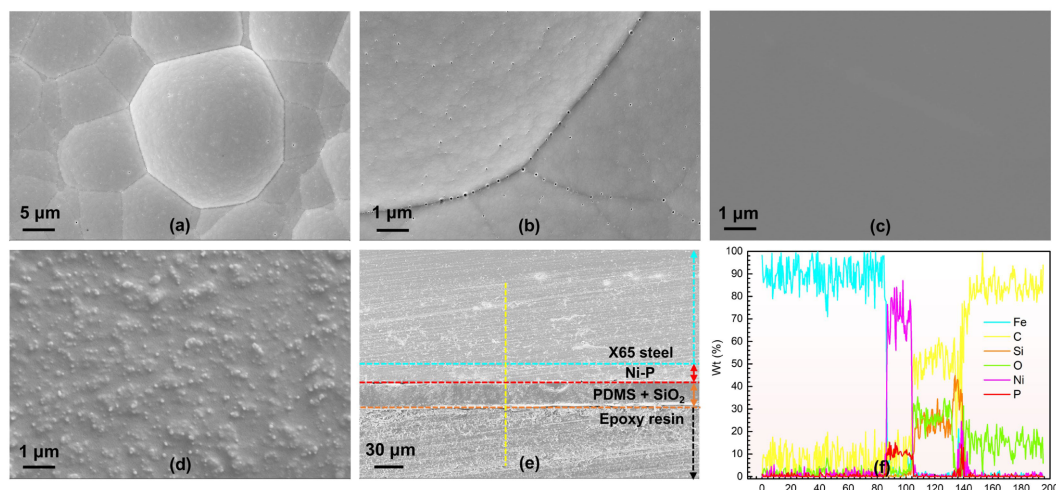


Fig. 2. Morphological and elemental characterization of the coatings. (a, b) Surface SEM images of the Ni-P interlayer at different magnifications, (c, d) Surface SEM image of the pure PDMS and PDMS-SiO₂ coatings, (e) Cross-sectional SEM morphology of the Ni-P/PDMS-SiO₂ composite coating on the X65 steel substrate, and (f) the corresponding EDS line scan profiles showing the elemental distribution across the multilayer structure (along the yellow dashed line in Fig. 2e. Note: The epoxy resin was used as the mounting material for cross-sectional observation).

ters of various sizes that were randomly yet densely embedded within the PDMS matrix. This combination resulted in a distinct micro-nano hierarchical roughness on the surface. This structure of SiO₂ clusters is expected to provide structural support for further modifications of the surface properties of smooth, pure PDMS.

To understand the internal structure and interface integrity of the prepared system better, cross-sectional morphology analysis was conducted via SEM (as shown in Fig. 2(e)). The micrograph clearly shows that a well-defined, dual-layer hierarchical structure has been formed on the X65 steel substrate. Notably, the interfaces of the X65 steel, the Ni-P interlayer, and the PDMS-SiO₂ topcoat were continuous and tightly bonded without any obvious microcracks, pores, or delamination. This showed good interfacial compatibility and structural stability. Furthermore, the thickness of each layer can be taken from the cross-sectional figure directly. The Ni-P underlayer was uniformly about 20 ± 2 μm , and the PDMS-SiO₂ superhydrophobic topcoat had a thickness of approximately 28 ± 3 μm . Based on a reasonable design, the total thickness of the composite coating was about 50 ± 5 μm , and thus a relatively physical barrier for limiting the penetration of corrosive media was formed.

The successful sequential construction of this multi-layer structure has also been verified by the corresponding EDS line scan profile (Fig. 2f), which was acquired along the cross-section (the yellow dashed line in Fig. 2e). The elemental distribution exhibited distinct and sharp transitions at the phase boundaries. Specifically, the innermost region had a prominent Fe peak and was thus identified as the bare X65 steel substrate. An abrupt emergence of Ni and P signals characterized the amorphous Ni-P sublayer. Subsequently, the outermost region was rich in high-intensity signals for Si, O and C, and thus SiO₂ nanoparticles were uniformly dispersed within the PDMS polymer matrix. These abrupt elemental changes at the interfaces of the respective layers confirmed the structural integrity of the composite coating, and a well-stratified, multi-tier defense

system was successfully built.

The crystalline structure of the Ni-P interlayer was characterized by XRD. As shown in Fig. 3a, a broad diffraction peak appeared at $2\theta \approx 44.5$, which was the typical characteristic of an amorphous structure. According to the EDS results, the formation of this amorphous phase was directly attributed to the oversaturated co-deposition of high content phosphorus atoms (13.3 wt.%, Fig. S1c), and it had the excellent corrosion resistance.

To analyze the chemical structure of the coatings, FT-IR spectroscopy was employed to examine the pure PDMS, SiO₂, and the PDMS-SiO₂ composite coatings. In Fig. 3b, the spectrum of the PDMS matrix was identified by the -CH₃ stretching vibration at 2960 cm⁻¹ and the sharp Si-CH₃ symmetric bending mode at 1260 cm⁻¹. In the range of 1010 cm⁻¹ and 1090 cm⁻¹, an asymmetric Si-O-Si stretching band was observed, confirming the formation of the siloxane network. For the bare SiO₂ fillers (Fig. 3c), typical signals for Si-O stretching (1080 cm⁻¹) and Si-O-Si bending (470 cm⁻¹) were clearly observed. After adding the nanoparticles to the polymer, the spectrum (Fig. 3d) still had the characteristic peaks of PDMS. A broadening and intensity increase of the Si-O-Si band were observed, indicating SiO₂ incorporation occurred. In addition, there was no significant -OH absorption band around 3400 cm⁻¹, thus the hydrophobic nature and low moisture sensitivity of the composite coating were also confirmed.

Measure the water contact angle (WCA) of the samples before and after 45 days of exposure to *P. aeruginosa* medium to determine their surface-wetting ability and extended stability (Fig. 3). At the beginning (day 0), the bare X65 steel was slightly hydrophilic and had a WCA of about 72° (Fig. 3e). The application of the Ni-P interlayer and the pure PDMS coating increased the initial WCAs to approximately 108° (Fig. 3f) and 124° (Fig. 3g), respectively, displaying intrinsic hydrophobicity. Notably, the Ni-P/PDMS-SiO₂ composite coating showed a very superhydrophobic state with an initial WCA exceeding 158° (Fig. 3h). This outstanding water-repellency was

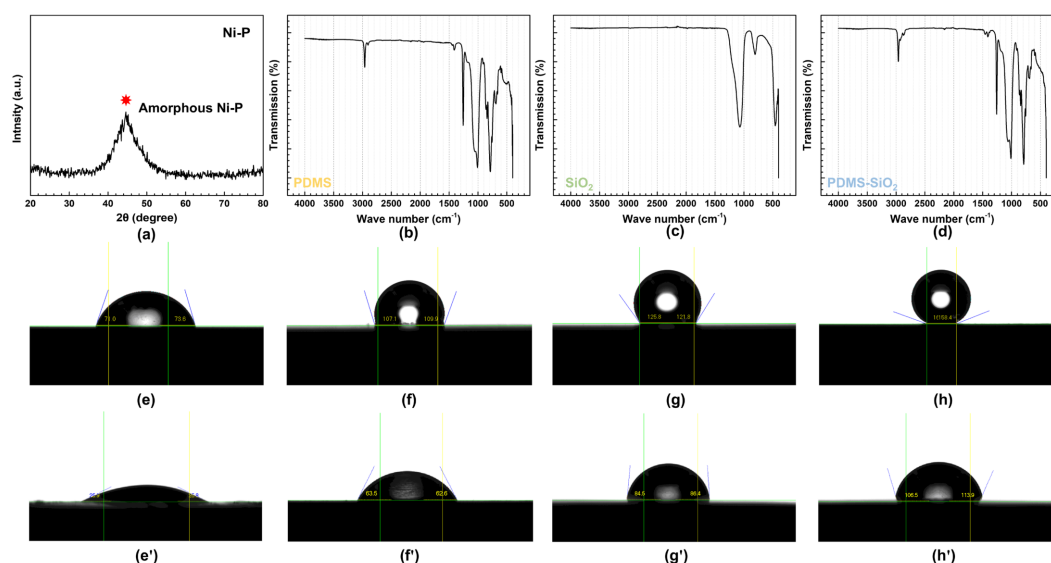


Fig. 3. Structural, chemical, and wetting characterization of the coatings. (a) XRD pattern of the electroless Ni-P interlayer; (b-d) FT-IR spectra of the PDMS, SiO₂ nanoparticles, and PDMS-SiO₂ composite coating; (e-h) Water contact angle (WCA) measurements at day 0 (initial) and after 45 days of immersion in the *P. aeruginosa* medium for (e, e') bare X65 steel, (f, f') Ni-P interlayer, (g, g') PDMS coating, and (h, h') Ni-P/PDMS-SiO₂ composite coating.

attributed to the hierarchical surface roughness provided by dispersed SiO_2 nanoparticles and the low surface energy of the PDMS matrix [31].

After 45 days of continuous immersion, all the samples exhibited various degrees of wetting state degradation due to prolonged hydrostatic pressure, gradual medium permeation, and the possible accumulation of microbial metabolites. The bare X65 steel (Fig. 3e') and the Ni-P interlayer (Fig. 3f') were severely hydrophilic, and their WCAs dropped sharply to around 25° and 63° , respectively. The main reasons for this substantial drop were that these unprotected areas accumulated a large amount of porous corrosion products and hydrophilic biofilms. Similarly, the WCA of the pure PDMS coating dropped to approximately 85° (Fig. 3g'), and it was likely that some polymer swelling and minor bacterial adhesion occurred, making it slightly more hydrophilic. Although the composite coating (Fig. 3h') showed a partial Cassie-to-Wenzel transition and a small decrease in its initial superhydrophobicity, it was still strongly hydrophobic and had a stable WCA of approximately 110° . The above hydrophobicity indicated that the composite structure can reduce the continuous inward diffusion of the corrosive bacterial medium.

The initial stage characteristics of MIC are that bacteria irreversibly adhere and subsequently form biofilms. Thus, using *P. aeruginosa* as the model microorganism, the anti-adhesion ability of the coating in the early stage was evaluated. As revealed by the fluorescence images in Figs. 4a and 4b, high-density green fluorescence was observed on both the bare X65 steel and Ni-P coating, indicating that bacteria were widely colonized and the biofilm was formed early. In contrast, the sur-

face coating based on PDMS significantly reduced the adhesion of bacteria. For the pure PDMS coating (Fig. 4c), the fluorescence intensity was significantly reduced, which was mainly attributed to the inherent low surface energy of the polymer matrix. Most notably, the Ni-P/PDMS- SiO_2 composite coating (Fig. 4d) exhibited anti-biofouling efficacy, with only a few fluorescent spots detected on the surface.

To quantitatively verify these visual observation results, the bacterial coverage area was calculated. Consistent with expectations, the bare X65 steel showed extensive bacterial adhesion in the initial stage, with a coverage area of approximately 5.2%. The hierarchical composite coating had a similar low bacterial coverage of only 0.05%. A large reduction in quantity (more than 98% compared to bare steel) shows that the hierarchical design successfully prevented *P. aeruginosa* attachment during the early settlement stage. Based on the wetting analysis, the superhydrophobic Ni-P/PDMS- SiO_2 surface maintained a stable air plastron in the Cassie-Baxter state, acting as a physical shield to prevent direct contact between the bacterial suspension and the solid surface.

The long-term protective performance of the coating was systematically evaluated over a 45-day test period, during which the sample was immersed in the *P. aeruginosa*-containing medium. As shown in Fig. 5a, the OCP of bare X65 steel was still at a very negative potential (around -700 mV vs. SCE), while the Ni-P/PDMS- SiO_2 composite coating had the most noble potential (near 100 mV), indicating a significantly reduced thermodynamic tendency for corrosion. As shown in Fig. 5b, the radius of the capacitive loop for bare X65 steel gradually increased over time, indicating a reduction in the corro-

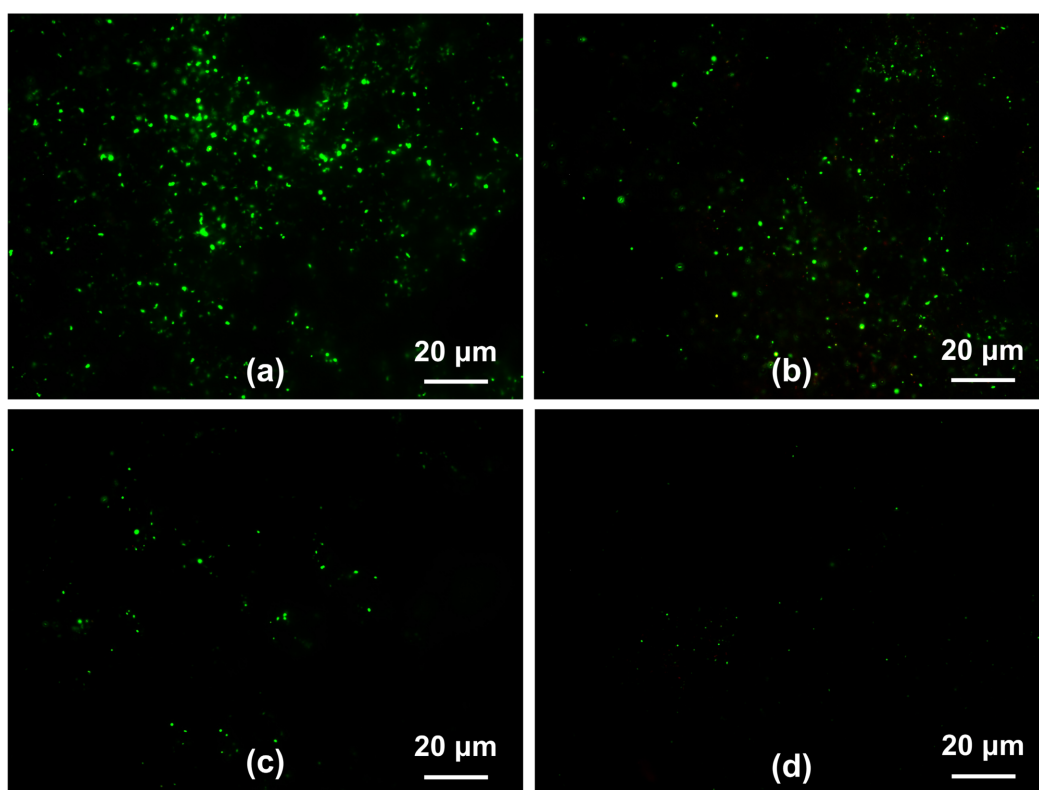


Fig. 4. Anti-bioadhesion performance against *P. aeruginosa*. Fluorescence microscopy images showing bacterial adhesion on different surfaces after 24 h of incubation: (a) bare X65 steel, (b) Ni-P interlayer, (c) Ni-P/PDMS coating, and (d) Ni-P/PDMS- SiO_2 composite coating. The green fluorescence represents live bacteria attached to the surfaces.

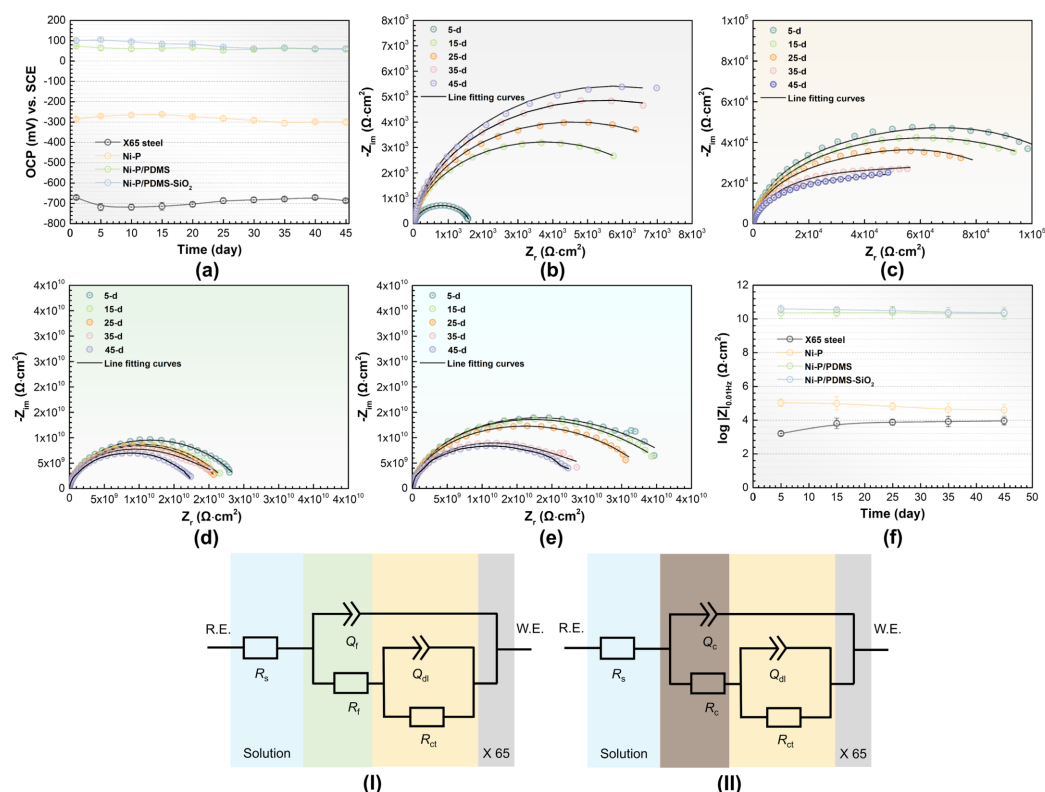


Fig. 5. Electrochemical stability of the samples during 45 days of immersion in *P. aeruginosa*-containing medium. (a) Open circuit potential (OCP) as a function of immersion time; (b–e) Nyquist plots of (b) bare X65 steel, (c) Ni-P interlayer, (d) Ni-P/PDMS coating, and (e) Ni-P/PDMS-SiO₂ composite coating at various intervals; (f) Evolution of the low-frequency impedance modulus ($|Z|_{0.01}$ Hz) derived from EIS data; Equivalent electrochemical circuits utilized for EIS data fitting: Model (I) for (b), Model (II) for both (c), (d) and (e).

sion rate. Based on bioelectrochemistry, this behavior was attributed to mass transport resistance. Long-term exposure led to an increase in the thickness of the biofilm matrix, trapping the insoluble iron corrosion products and forming a heterogeneous biomineralized layer. However, despite the retarding effect of this biomineralized layer, corrosion of the bare steel still occurred continuously. Compared with the bare substrate, the single Ni-P interlayer (Fig. 5c) provided a moderate barrier protection, but its corrosion resistance gradually declined during the early immersion stage. Therefore, it was concluded that the electrolyte diffused through a micro-crack in the Ni-P layer. Notably, in the mid-to-late stages of immersion, the variations in the Nyquist loops for the Ni-P coating stabilized, suggesting that the interface reached a quasi-stable state after initial corrosion. For the pure PDMS coating (Fig. 5d), the diameters of the Nyquist semicircles remained relatively constant during the entire immersion period and thus exhibited a stable barrier effect and chemical stability of the PDMS polymer.

The EIS data obtained were fitted with the Model I and Model II. R_s is the solution resistance, R_f , R_c and R_{ct} are the film, coating, and charge transfer resistances, respectively, and Q_f , Q_c and Q_{dl} are the corresponding constant phase elements for the film, coating, and electrical double-layer capacitances. The fitting parameters are shown in Table 1 and Table 2, respectively. Changes in the above individual factors provide a specific reason for the performance decline. R_c is the anti-permeation capability of the coating in an electrolyte, and Q_c is a highly sensitive index of water absorption [32]. The Ni-P/PDMS-SiO₂ composite had a very high R_c value and a low Q_c value simultaneously. Thus, the multi-layer design could not allow corrosive media or bacterial metabolites to penetrate physically. A highly repellent PDMS-SiO₂ topcoat was used to reduce the liquid-solid contact area, and a dense, amorphous Ni-P sublayer effectively blocked the ion diffusion path. In addition, the large R_{ct} and small Q_{dl} of the composite showed that the electrochemical corrosion kinetics at the underlying X65 steel interface was almost entirely inhibited.

Table 1. Best-fit parameters of the equivalent circuits for bare X65 steel after different immersion times in the *P. aeruginosa*-containing medium.

Incubation (Day)	R_s ($\Omega \cdot \text{cm}^2$)	Y_f ($\Omega^{-1} \cdot \text{cm}^{-2} \text{ s}^n$)	n_1	R_f ($\Omega \cdot \text{cm}^2$)	Y_{dl} ($\Omega^{-1} \cdot \text{cm}^{-2} \text{ s}^n$)	n_2	R_{ct} ($\Omega \cdot \text{cm}^2$)
X65 steel							
5	27.3	2.24×10^{-4}	1	11.5	5.65×10^{-4}	0.92	1590
15	26.7	8.21×10^{-4}	0.94	59.6	1.85×10^{-3}	0.75	1581
25	26.9	5.15×10^{-4}	1	17.8	3.48×10^{-4}	0.81	9293
35	26.6	5.35×10^{-4}	1	9.2	4.22×10^{-4}	0.85	11021
45	27.0	4.96×10^{-4}	1	8.4	3.95×10^{-4}	0.84	12385

Table 2. Best-fit parameters of the equivalent circuits for various coatings (Ni-P coating, Ni-P/PDMS coating, and Ni-P/PDMS-SiO₂ composite coating) after different immersion times in the *P. aeruginosa*-containing medium.

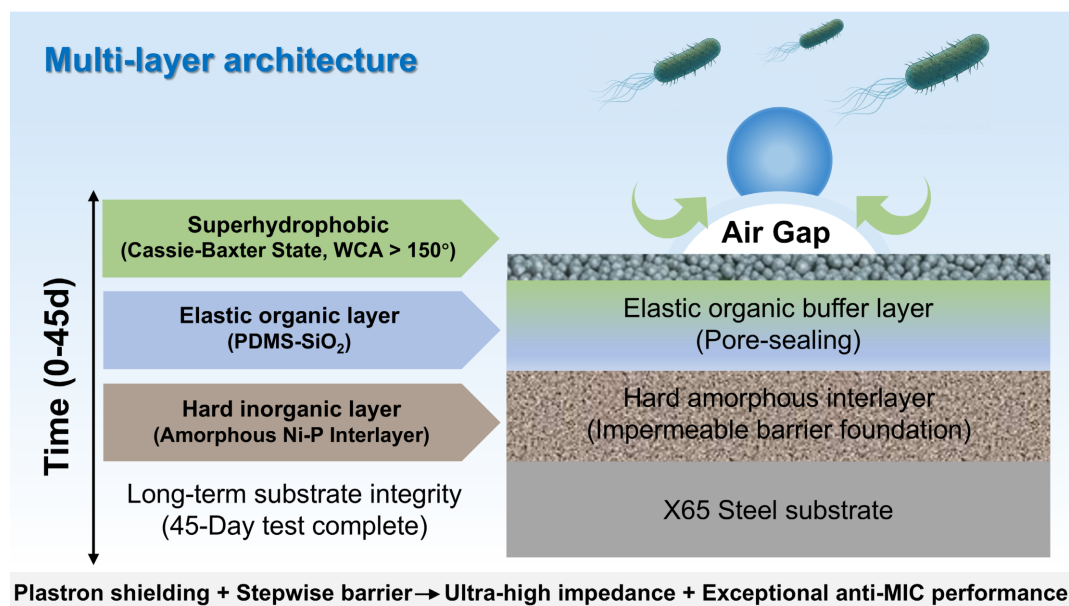
Incubation (Day)	R_s ($\Omega\cdot\text{cm}^2$)	Y_c ($\Omega^{-1}\cdot\text{cm}^{-2}\text{ s}^n$)	n_1	R_c ($\Omega\cdot\text{cm}^2$)	Y_{dl} ($\Omega^{-1}\cdot\text{cm}^{-2}\text{ s}^n$)	n_2	R_{ct} ($\Omega\cdot\text{cm}^2$)
Ni-P coating							
5	26.11	2.68×10^{-5}	0.93	8.34×10^4	7.37×10^{-5}	0.58	7.37×10^4
15	25.86	2.66×10^{-5}	0.94	6.37×10^4	5.86×10^{-5}	0.66	6.76×10^4
25	25.91	2.67×10^{-5}	0.93	6.04×10^4	7.44×10^{-5}	0.53	5.49×10^4
35	26.20	3.88×10^{-5}	0.92	3.46×10^4	7.37×10^{-5}	0.56	1.43×10^4
45	24.89	4.09×10^{-5}	0.92	2.76×10^4	8.42×10^{-5}	0.55	1.01×10^4
Ni-P/PDMS coating							
5	0.008	4.46×10^{-11}	0.96	1.24×10^{10}	7.31×10^{-11}	0.56	1.31×10^{10}
15	0.008	4.47×10^{-11}	0.97	1.06×10^8	1.96×10^{-11}	0.46	2.30×10^{10}
25	0.009	4.43×10^{-11}	0.97	9.54×10^7	1.95×10^{-11}	0.45	2.39×10^{10}
35	0.010	4.54×10^{-11}	0.97	7.21×10^7	2.31×10^{-11}	0.44	1.95×10^{10}
45	0.091	4.73×10^{-11}	0.96	1.17×10^7	1.48×10^{-11}	0.46	1.35×10^{10}
Ni-P/PDMS-SiO₂ composite coating							
5	0.007	3.55×10^{-11}	0.98	2.12×10^{10}	8.06×10^{-11}	0.51	2.52×10^{10}
15	0.006	4.91×10^{-11}	0.97	1.89×10^{10}	7.45×10^{-11}	0.58	2.31×10^{10}
25	0.009	3.94×10^{-11}	0.97	1.35×10^{10}	6.27×10^{-11}	0.57	2.28×10^{10}
35	0.009	4.61×10^{-11}	0.95	1.25×10^{10}	2.73×10^{-11}	0.59	1.46×10^{10}
45	0.010	4.54×10^{-11}	0.96	1.21×10^{10}	1.40×10^{-11}	0.62	1.33×10^{10}

The low-frequency impedance modulus ($|Z|_{0.01\text{ Hz}}$) can be employed as an overall protective efficiency indicator for coating systems. As shown in Fig. 5f, the $|Z|_{0.01\text{ Hz}}$ values of the various samples had different steps of change over the 45-day immersion period. The bare X65 steel consistently had the lowest impedance value ($10^3 \sim 10^4 \Omega\cdot\text{cm}^2$). The single Ni-P interlayer and Ni-P/PDMS coating achieved leaps to magnitudes of $10^5 \Omega\cdot\text{cm}^2$ and $10^{10} \Omega\cdot\text{cm}^2$, respectively. Notably, the impedance of the Ni-P/PDMS-SiO₂ composite coating was about $10^{10.5} \Omega\cdot\text{cm}^2$, taking the largest Nyquist loop radius among all the systems. Even in the mid-to-late stage of immersion, when a small drop in R_c and R_{ct} occurred, the overall $|Z|_{0.01\text{ Hz}}$ was still relatively high at the $10^{10} \Omega\cdot\text{cm}^2$ level and significantly larger than that of other samples.

According to the changes in impedance, many levels of

shielding was achieved incrementally. A Ni-P layer was added to provide a base for the first physical cover, and PDMS was used to seal the pores through low-surface-energy properties. The superhydrophobic structure of SiO₂ nanoparticles also formed an insulating barrier in the Cassie-Baxter wetting state by trapping a stable air gap at the solid-liquid interface. This increased the R_{ct} , inhibited the adsorption of *P. aeruginosa* on the surface, and had a long-term protective effect.

Based on the electrochemical and morphological analyses, the comprehensive protective mechanism of the hierarchical Ni-P/PDMS-SiO₂ coating is characterized by a multi-layer protection system (Fig. 6). The total impedance modulus of this system remained stable within 45 days, approximately $10^{10} \Omega\cdot\text{cm}^2$. The primary shield was the Cassie-Baxter air plastron trapped within the micro/nano-structured superhydrophobic

Fig. 6. Schematic illustration of the defense mechanism of the Ni-P/PDMS-SiO₂ composite coating against *P. aeruginosa* induced MIC.

topcoat. This plastron acted as a physical barrier that reduced the effective solid-liquid contact area. Furthermore, corrosive species that penetrate this first shield were also intercepted by the underlying barrier layers. The elastomeric PDMS-SiO₂ buffer layer sealed the pores of the metallic interlayer and accommodated mechanical stresses. Below this was a dense amorphous Ni-P interlayer that prevented the diffusion of residual corrosive species to the X65 steel substrate. The hierarchical protection mechanism reduced the contact area of corrosive media through the air barrier layer and prevented residual substances from penetrating by the dense underlying layer. As a result, when immersed in the *P. aeruginosa* medium for a long time, the corrosion resistance was increased by six orders of magnitude, and the base integrity is maintained.

4. Conclusion

In short, a multi-layered Ni-P/PDMS-SiO₂ composite coating with superhydrophobic properties was successfully prepared on X65 steel and showed good durability against MIC. Electrochemical results showed that the composite coating reached an ultra-high impedance plateau of about 10¹⁰ Ω·cm², and kept a stable protective barrier for 45 days. This was a leap of six orders of magnitude in resistance compared with bare X65 steel (10³ ~ 10⁴ Ω·cm²), thereby better protecting the substrate from the corrosive environment.

The above protection was achieved by a dual-function system, that is, the superhydrophobic topcoat significantly reduced the solid-liquid contact area. This air plastron prevented the adhesion of *P. aeruginosa*. Meanwhile, an amorphous Ni-P interlayer was added to reduce the permeability of residual corrosive species to the substrate, while a PDMS matrix was used for pore sealing. The hierarchical superhydrophobic structure serves as a relatively reliable and long-lasting protective measure for industrial facilities in environments with a high density of microorganisms.

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CRedit authorship contribution statement

Zhaoqi Chen: Experimental operation, Data curation, Writing-original draft; **Yanan Pu:** Writing-review & editing; **Xuwen Cao:** Writing-review & editing; **Hongbo Zeng:** Resources, Supervision, Writing-review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The data that support the results of this study are available from the corresponding author on reasonable request.

Electronic Supplementary Material (ESM)

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