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Fabrication of Self-Healing Superhydrophobic Coatings on Steel as Effective Corrosion Barrier

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Abstract: *It has been demonstrated that silica-based coatings prepared using sol-gel technology are highly chemically stable and reduce metal corrosion, making them ideal for applications such as protecting metals in marine industries. Moreover, this technology offers an eco-friendly way to produce surface coatings, potentially replacing toxic pre-treatment coatings of traditional chromate. This study aims to produce superhydrophobic silica coatings to protect the steel substrate from corrosion. The approach includes a modification of base* SiO_2 *coating by 1H, 1H, 2H, 2H-perflurodecyltriethoxysilane (FAS) and its effect on wetting and corrosion resistive behavior of coatings. X-ray photoelectron spectroscopy (XPS) characterized the surface elemental composition of coatings. Water contact angle (WCA) and atomic force microscopy (AFM) are utilized to determine coatings' wetting behavior. The corrosion protection potentials of coatings were examined by potentiodynamic polarization curve (PDPC); all these are supported by surface morphology analysis using scanning electron microscopy(SEM). The results of PDPC show an increase in corrosion protection due to the modification of base silica coating by FAS. Additionally, modified coatings exhibited improved resistance to cracking and self-cleaning ability.*

Keywords: Superhydrophobic, Sol-gel method, Dip Coatings, Anticorrosive, self-cleaning

I. INTRODUCTION

There has been substantial scientific attention paid to superhydrophobic coatings [1] in a variety of fields, including self-cleaning coatings [2], antifog [3], anti-biofouling [4], anticorrosion [5-8], selective transport of microdroplets [9] and oil/water separation [10]. Coatings are said to be superhydrophobic if the contact angle is greater than 150^0 and the Sliding angle is less than $5\text{-}10^0$. The low surface energy and micro-nano surface roughness are responsible for the wetting behavior of these coatings. Therefore, superhydrophobic coatings can be achieved by chemically treating surfaces with low-surface energy materials or by creating micro-nano roughness on low-energy surfaces.

There has been substantial scientific attention paid to superhydrophobic coatings [1] in a variety of fields, including self-cleaning coatings [2], antifog [3], anti-biofouling [4], anticorrosion [5-8], selective transport of microdroplets [9] and oil/water separation [10]. Coatings Researchers have recently devoted a great deal of effort to preparing hierarchical structures using a variety of methods, including electrochemical deposition [5,11], chemical etching [13,14], chemical vapor deposition [14,15], and template-based embossing and lithography [16.17]. Nevertheless, these methods require highly advanced instrumentation and multiple steps at high temperatures. The coatings made by these approaches have a high cost and show weak adhesion, poor durability, and transparency. The durability of superhydrophobic coatings is rapidly affected by harsh conditions, viz humid or wet environments, exposure to strong acid or base, and UV radiation, leading to a short lifetime.

Optically transparent superhydrophobic coatings have great applications in self-cleaning glasses used in solar panels, windows of skyscrapers, and optical devices. Nevertheless, most artificial superhydrophobic surfaces lack the transparency needed for self-cleaning coatings. However, creating optically transparent superhydrophobic coatings takes work, as optical transparency and surface roughness are inversely related. It is seen that surfaces with roughens greater than 400 nm scatter the light [18,19]and lower the optical transparency considerably.

Along with optically transparent self-cleaning coatings, anticorrosive coatings that protect metal surfaces from harsh environment are also focused on in the last decade. The most extensively used method for shielding metal surfaces from corrosion is coating them with protective layers. Coating metal surfaces is crucial for enhancing their surface properties and expanding their functionalities. As a result, numerous research activities have been started to create hybrid coatings systematically to produce a new class of materials with a greater performance [20, 21] by combining various materials with complementary properties to meet the primary requirements. [20, 22, 23] To achieve effective anti-corrosion coatings, the protective film must possess both adhesive and hydrophobic properties. [24] The two features are, however, only sometimes compatible. However, during actual use, the coatings are easily impacted by the environment, which inexorably results in the formation of cracks. The corrosive substance can easily reach the underlying substrate through these cracks and erode them. With no manual intervention, the corrosion resistivity of the coatings decreases gradually. Furthermore, these microcracks cannot be repaired effectively using conventional welding and repair methods. The researchers sought to overcome these limitations by drawing inspiration from naturally efficient coatings on creatures. They discovered that these coatings restore protection by healing skin wounds. Therefore, smart corrosion resistive coatings have been developed by using the idea of self-healing.

According to studies, the anti-corrosion properties of coatings can be enhanced by improving the passive barrier effect of coatings by adding a healing agent to the organic coating matrix. [25-27] These anticorrosive coatings can restore or even improve their anti-corrosion properties with minimal manual intervention. [28-30] This is because the healing agent, i.e., the inhibitor carried in the organic matrix, fills the crack before corrosion starts. Inhibitor reduces the rate of corrosion by preventing anodic and cathodic reactions. The corrosion inhibitor is a chemical component, the addition of which, in a very small quantity to the metal environment, decreases, controls, and inhibits corrosion. To achieve long-term durability, it would be best to fabricate superhydrophobic surfaces with self-healing abilities. Rao et al. fabricated superhydrophobic coatings with self-healing abilities using fluoroalkyl silane (FAS) [25]. If the top layer of coatings is damaged, the restoration process is accelerated due to the preserved FAS that migrates to the surface to heal the damaged surfaces. Zhang et al. fabricate coatings that self-heal the cracks on heating.[31]

II. MATERIALS AND METHODS

2.1 Materials

Commercially available still alloy size of 1.00 cm × 4.00 cm is ideal metal substrate for coatings. Propyltrimethoxysilane(PTMS), Polydimethylsiloxane polymer (PDMS), and 1H,1H,2H,2H-perfluorodecyltriethoxysilane (FAS) were obtained from Sigma Aldrich. The common chemicals, ammonia, ethanol, and acetone were purchased from Lobachem. All starting materials were used as received.

2.2 Fabrication of superhydrophobic coatings

Strips of steel substrate were mechanically polished with emery paper grade 600, degreased with water, and finely with acetone. The SiO_2 coating solution was prepared by mixing the PTMS, ethanol, and distilled water in the ratio 2.5/45/2/5 %v/v, respectively. After magnetically stirring for 15 minutes, a 2.5%v/v of PDMS was added to enhance the solution. To complete hydrolysis and condensation, 0.5 N HNO_3 was added dropwise to the above solution, and the pH of the solution was maintained at 4. The solution was then magnetically stirred at 45^0 C for 4 h. This solution is served as solution 'A'. Meanwhile, a 0.4 mM solution of FAS in ethanol was fabricated. This solution is served as solution 'B'. Solution B is mixed with part of solution A and magnetically stirred for another 4 h at 45^0 C. This solution served as a solution C.

After the sol was prepared, cleaned steel and copper substrates were dipped vertically into sol A and Sol C. The bonding of silane molecules onto the metal surface is fast due to rapid hydrogen bonding between SiOH and the metal hydroxyl groups. [32-34] However, a short submersion time may result in inhomogeneity in the coating coverage [35]. Therefore, the substrate was kept in a sol for a sufficient time and withdrawn at a constant rate of 2 mm per second to get the desired uniform thickness of the coatings. After removing from the sol, coatings are allowed to dry at room temperature for 1h and then heat treated at 100^0 for 2 h. The coatings of sol A named SiO_2–steel coatings, whereas the coatings of sol C are named FAS-SiO_2-steel coatings.

III. CHARACTERIZATION

The surface elemental composition of the coated stainless steel was characterized using X-ray photoelectron spectroscopy (XPS). XPS measurements were performed using a PHI 5000 Versa Probe II XPS system (ULVAC-PHI, Japan). Monochromatic Al Kα excitation (1486.6 eV) at 100 W, a pass energy of 46.95 eV, and a step size of 0.05 eV were used to record the XPS spectra. Contact angle measurements of coatings were performed by putting 10 µl of distilled water drop on the coating surface at room temperature. For this standard goniometer (Ramehart Instrument Co., USA) equipped with a CCD camera was used. The surface topography of the coatings was studied using Atomic Force Microscopy (AFM Nanoscope E' of 'Digital Instruments, USA). Potentiodynamic polarization measurements were carried out to test the corrosion resistance abilities of the coatings. All measurements were performed using an electrochemical analyzer (CHI604E, China) in 3.5wt.% NaCl solution at room temperature with three electrode methods. Scanning Electron Microscopy (SEM) was used to examine the coatings' surface morphology and homogeneity (Model: JEOL-JSM-6360).

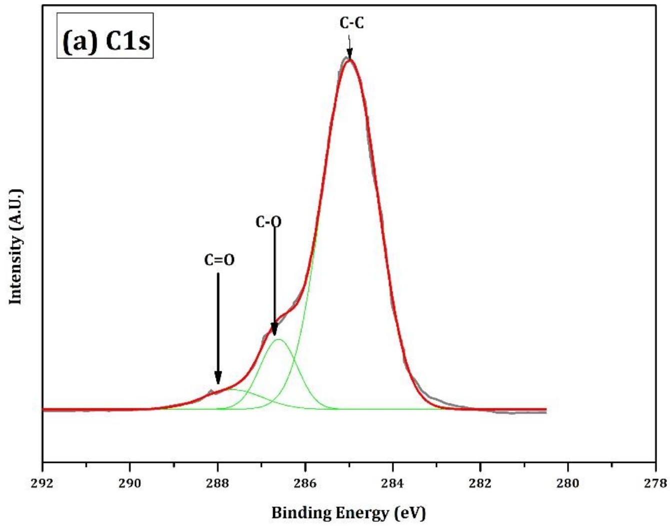
IV. RESULTS AND DISCUSSION

4.1 X-ray photoelectron spectroscopy (XPS)

The surface elemental composition of the pristine steel, SiO_2-steel, and FAS-SiO_2-steel Coatings was characterized by the XPS to determine the presence of fluoroalkyl group and silica shells in the coatings whose migration confers superhydrophobicity. For this data were obtained for C1s (pristine steel), C 1s, O 1s, and Si 2p (SiO_2-steel) and C 1s, F 1s, and Si 2p (FAS-SiO2-steel). Fig. 1a, b, and c Show the C 1s core level spectra for pristine steel, C 1s core level spectra and corresponding O 1s and Si 2p spectra in insets for SiO_2- steel, and C1s core level spectra and corresponding F 1s and Si 2p spectra in insets for FAS-SiO_2-steel coatings. Before modification, pristine steel C 1s spectra were deconvoluted into three peaks at binding energies 285, 286.6, and 287.7 eV attributed to C-C, C-O, and C=O, respectively. After coatings with SiO_2, SiO_2-steel coatings C 1s spectra could be resolved into three peaks at binding energies of 283.1, 284.7, and 286.6 eV corresponding to C-Si, C-C, and C-O, respectively. [36-38] The C-Si and C-C peaks could be linked to SiO_2. The modified FAS-SiO2-steel coatings C 1s spectra were deconvoluted into seven peaks at binding energies 283.9, 285, 286.8, 288.1, 290.4, 292.1, and 293.4 eV attributed to C-Si, C-C, C-O, C=O, CF_2-CH_2, CF_2-CF_2, and CF_3-CF_2 respectively. [39-41] The result conforms to the presence of FAS on the surface of modified coatings.

In the deconvolution of C 1s spectra of SiO_2- steel coatings, the surface carbon and βSiC could be linked to the C-C and Si-C compounds, respectively. The bond present in popyltrimethoxysilane may be responsible for the C-O compound. The O1s XPS peak at 533.15 could be related to SiO_2 production by the popyltrimethoxysilane. The Si 2p XPS spectrum of SiO2-steel coatings is dominated by a peak at 103.56 eV could be attributed to $Si4^+$ in SiO_2.[38]

The C 1s XPS spectra of FAS-SiO_2-steel deconvolute into seven peaks. The CF_2-CH_2, CF_2-CF_2, and CF_3-CF_2 bonds correspond to existing bonds in FAS which lowers the surface energy of coatings and is responsible for higher WCA. The high-resolution F 1s spectra show an intense peak at 690.6 eV conforms to the presence of Florine at the surface in FAS-SiO_2-steel.



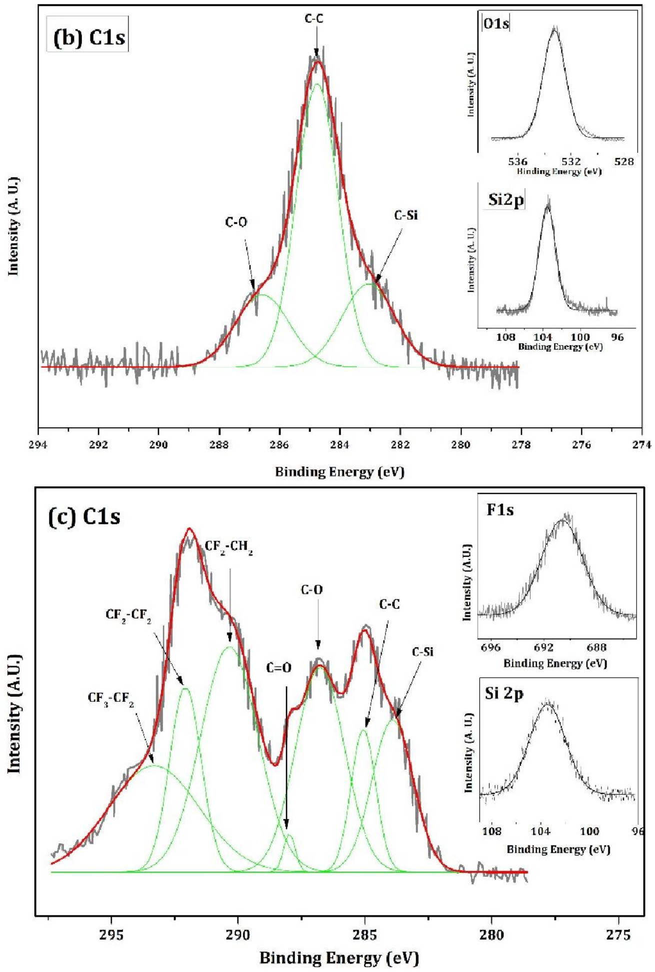


Fig. 1 Deconvolution of C 1s peaks of (a) pristine steel, (b) SiO_2-steel and (c) FAS- SiO_2-steel

4.2 Contact angle measurement

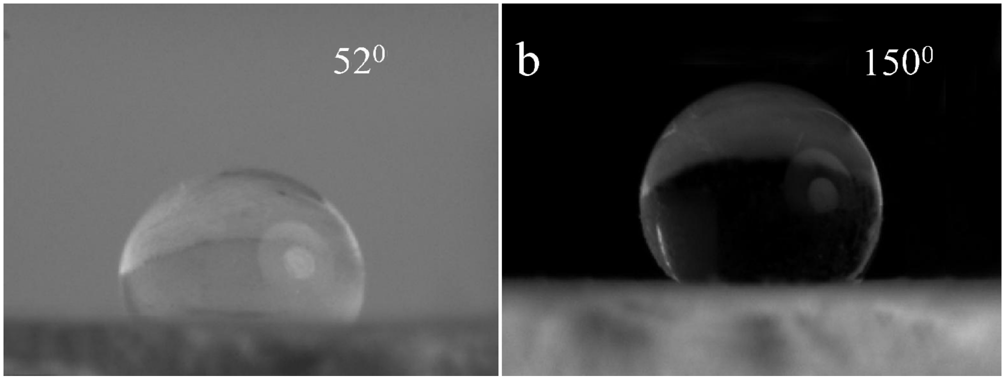


Fig. 2 Water contact angle measurement for (a) SiO_2-steel and (b) FAS- SiO_2-steel

Static WCA measurements explored the wetting behavior of pristine steel, SiO_2-steel, and FAS-SiO_2-steel coatings. Fig. 2a and b show the images of water droplets SiO_2-steel and FAS-SiO_2-steel coatings. A static WCA of $110^0 \pm 2^0$ and $150^0 \pm 2^0$ was observed for the SiO_2-steel and FAS-SiO_2-steel coatings, respectively. The SiO_2-steel coatings showed a high-water contact angle compared to pristine steel (not shown in Fig.). This ensures that coating a steel substrate with a silane layer makes the metal surface hydrophobic. However, an additional increase in WCA was observed for FAS-SiO_2-steel coatings. This signifies that water adsorption could be greatly diminished on FAS-SiO_2-steel coatings favoring its stability increase in different harsh conditions. This high WCA of FAS-SiO_2-steel coatings is attributed to the presence of fluorine, well known for lowering the surface free energy. Fluorine forms a stable covalent bond with carbon due to its small atomic radius and highest electronegativity among all atoms.

4.3 Atomic Force Microscopy (AFM)

The wettability of coating surfaces mainly depends upon two factors, surface chemical composition, and surface roughness. Fig. 3a, b, and c show the standard 3D topography images and roughness profile of pristine steel, SiO_2-steel, and FAS- SiO_2-steel, respectively. The images were recorded at 2×2 μm^2 planar in contact mode. It was found that the pristine steel sample is smoother than that coated steel samples. After coating with SiO_2 and FAS-SiO_2 overall surface roughness of the coatings was examined. The root means square (RMS) roughness value of the SiO_2-steel and FAS-SiO_2-steel increases significantly compared to pristine steel. The surface roughness of pristine steel is 4.9 A^0, whereas that of SiO_2-steel and FAS- SiO_2-steel coatings are 31.07 nm and 54.57 nm, respectively. The increase in WCA for FAS- SiO_2-steel coatings as compared to SiO_2-steel is partly attributed to this increase in surface roughness. The measure cause of an increase in WCA is low surface energy Florine-Carbon species at the surface of FAS- SiO_2-steel coatings.

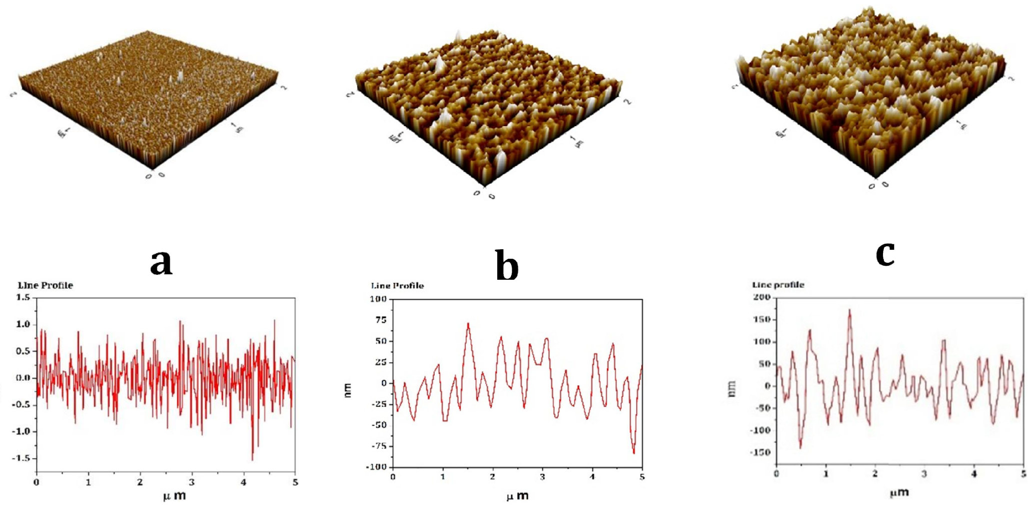


Fig. 3 3D AFM images and roughness profile of (a) pristine steel, (b) SiO_2-steel and (c) FAS- SiO_2-steel

4. 4 Potentiodynamic Polarization Curve

The anti-corrosion behavior of the three samples viz pristine steel, SiO2-steel, and FAS-SiO2- steel were demonstrated based on their electrochemical measurements. The potentiodynamic polarization (PDS) method was implemented to check the protective abilities of the SiO_2-steel, and FAS-SiO_2- steel coatings. Fig. 4 shows the potentiodynamic polarization curve for the different samples.

All samples were examined using a three-electrode system at room temperature after being submerged in 3.5 wt.% NaCl aqueous solution for 48 hours. A coating sample, a saturated calomel electrode, and a platinum electrode were the three electrodes used in the measurement as a working electrode, reference electrode, and auxiliary electrode, respectively. The polarization measurements were carried out at a scanning rate of 10 mV/s and the potential varies from -1 to -0.1 V.

Table 1 shows the results of potentiodynamic polarization measurements, which include corrosion potential (E_{corr}), corrosion current density (j_{corr}), and anodic and cathodic Tafel constants (βa, βc). As per the Tafel plot in fig. 4 and the corresponding results in table 1, the cathodic and anodic branches shifted to lower current densities. The corrosion current density j_{corr} reduced for SiO2-steel (5.04 × 10-6A/m2) and FAS-SiO2-steel (1.28 × 10-7 A/m2) coatings as compared to pristine steel (9.24 × 10-5A/m2) sample. The FAS-SiO2-steel coatings show the lowest j_{corr}. Tafel plot shows that E_{corr} shifts towards more positive potential in the order pristine steel < SiO2- steel< FAS-SiO2-steel. E_{corr} increased to -0.47 V and -0.43 V from -0.62 V for pristine steel after being coated with SiO2 and FAS-modified SiO2, respectively. This significant increase in E_{corr} and decrease in j_{corr} conforms to the protection of steel substrate from corrosion. This positive shift in E_{corr} indicates more anodic inhabitation than the cathode, corresponding to fluorine-carbon atoms bridging to the substrate. [42,43] A similar trend was found in the Tafel constant βa and βc. Compared with pristine steel, SiO2-Steel, and FAS-SiO2- Steel showed a rising trend in Tafel constants, as shown in Table 1.

Sample	**E_{corr} (V)**	**j_{corr} (A/m^2)**	**β_a (V)**	**β_c (V)**
Pristine Steel	- 0.62	9.24×10^{-5}	0.099	0.025
SiO2-Steel	- 0.47	5.04×10^{-6}	0.20	0.13
FAS-SiO2-Steel	- 0.43	1.28×10^{-7}	0.21	0.31

Table 1 Parameter obtained from Tafel extrapolation for pristine steel, SiO_2-steel, and FAS- SiO_2-steel

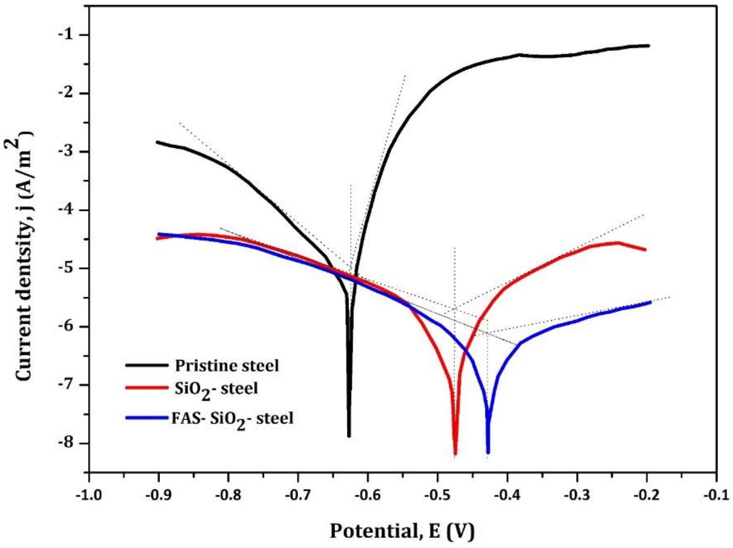


Fig. 4 Potentiodynamic polarization curve for pristine steel, SiO_2-steel, and FAS- SiO_2-steel

4.5 Scanning Electron Microscopy (SEM)

Both coated samples, SiO2-steel and FAS-SiO2-steel, showed the potential to provide high resistance to corrosion during immersion. Both coatings appeared intact after immersion, as observed by visual examination. Apparently, neither coating had been degraded or damaged immediately after immersion. However, for long immersion times, over 15 days, the SiO2-steel coatings developed micro cracks. Fig. 6a and c show the SEM images of the SiO2-steel coatings after 15 days of immersion in 3.5% NaCl solution. Coatings show cracks of 5 to 10 µm wide on the surface. The water underneath the film may cause swelling and the breakdown of coating-substrate adhesion. This breakdown of coating-substrate adhesion could lead to the cracking observed in SiO2-steel.[42] Despite these conditions, FAS-SiO2-steel exhibits outstanding crack resistance, reflecting the flexibility of the modified coatings, as shown in Figures 6b and d.

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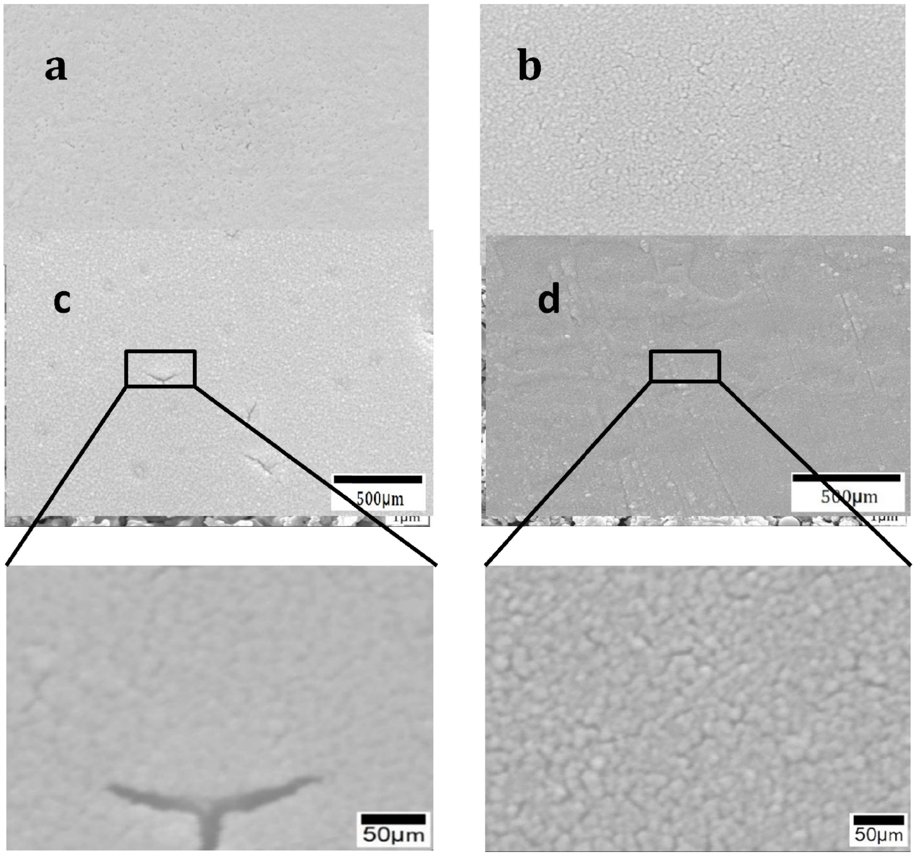

Fig. 5 SEM micrographs of a SiO_2-steel and FAS- SiO_2-steel immediately after immersion(a,b) and after immersion for 15 days (c, d) respectively.

V. CONCLUSION

The addition of FAS precursor improved the corrosion protection abilities of base SiO2 coatings. The electrochemical experiments for corrosion testing confirm the high corrosion resistance, and wetting measurements confirm higher WAC for FAS-modified coatings. Additionally, a fluorinated group from FAS in FAS-SiO2-steel coatings exhibits resistance against cracking for long immersion in corrosive liquid. So, exploring the hydrophobic properties of FAS in low concentrations will be worthwhile for applications that require self-cleaning, anti-corrosive, and anti-icing properties.

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