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ARTICLE

Synergistic Enhancement of Corrosion Resistance and Electrical Conductivity of TaTiN Coating for Ti Bipolar Plates

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Abstract: Coatings with strong corrosion resistance and high electrical conductivity on bipolar plates are urgently required for large-scale applications of proton exchange membrane water electrolysis (PEMWE). High-performance TaTiN coatings were fabricated via magnetron sputtering. Results show that TaTiN coatings with preferred (111) planes and refined grains show higher corrosion resistance and better electrical conductivity. On the one hand, the closely-packed (111) plane favors a dense texture; moreover, grain refinement can smooth the surface and facilitate the formation of a continuous, compact passive layer, thereby improving corrosion resistance. On the other hand, the small lattice mismatch between TaTiN (111) and Ti (0001) enhances interfacial bonding and thus electrical conductivity across the interface, resulting in lower interfacial contact resistance (ICR). The optimized TaTiN coating shows a low corrosion current density of $6.56 \times 10^{-7} \text{ A} \cdot \text{cm}^{-2}$ and an ICR of $9.11 \text{ m}\Omega \cdot \text{cm}^2$, indicating the strong potential of TaTiN coatings to protect metallic bipolar plates in PEMWE systems.

Key words: TaTiN coating; corrosion resistance; electrical conductivity; bipolar plates; proton exchange membrane water electrolysis

1 Introduction

Hydrogen energy represents a promising candidate for clean energy applications, owing to its high energy density and zero carbon emissions. Proton exchange membrane water electrolysis (PEMWE) is a key technology for hydrogen production via renewable energy, capitalizing on its excellent dynamic response and broad load adaptability^[1-4]. Bipolar plates (BPs), a key component of PEMWE cells, conduct current, provide structural support, and remove residual water^[5-8]. Typically, BPs operate under harsh conditions, including strongly acidic electrolytes (pH<3), high potentials (1.5–2 V), and elevated temperatures (70–80 °C). Consequently, BPs must exhibit both high corrosion resistance and excellent electrical conductivity. Surface modification is a main strategy to enhance these two properties^[9-11]. Precious metal coatings

(Pt, Ag, Au, etc) can protect Ti BPs because of their outstanding corrosion resistance and electrical conductivity. Nevertheless, their high cost has spurred the search for alternative materials, leading to considerable attention to transition metal nitrides^[12-17]. For example, Choe et al^[18] prepared TaN_x coatings on 316L stainless steel by magnetron sputtering, with a low corrosion current density (I_{corr}) of $0.333 \mu\text{A} \cdot \text{cm}^{-2}$ and a low interfacial contact resistance (ICR) of $11 \text{ m}\Omega \cdot \text{cm}^2$. Sun et al^[9] prepared a single δ -NbN coating on Ti BPs by magnetron sputtering, with I_{corr} down to $0.011 \mu\text{A} \cdot \text{cm}^{-2}$. However, the ICR values are still higher than the U. S. Department of Energy requirement ($\text{ICR} < 10 \text{ m}\Omega \cdot \text{cm}^2$)^[19]. TaN and TiN have the same crystal structure and similar lattice constant; as a result, Ta and Ti can replace each other in the TaTiN solid solution^[20]. This intrinsic compatibility offers two key advantages for the TaTiN coatings on Ti substrates. First,

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the small lattice mismatch between TaTiN and Ti substrates enhances the bonding strength of the TaTiN coatings on Ti substrates^[21], improving interfacial electrical conductivity and reducing the ICR value. Second, this multi-component structure can refine the grains and promote the formation of a continuous, compact passive layer, improving corrosion resistance.

In this study, TaTiN coatings were fabricated on Ti substrates by magnetron sputtering. The N₂/Ar flow ratio, deposition pressure, and substrate temperature were optimized; the electrical conductivity, corrosion resistance, and hydrophobicity were evaluated. Accordingly, the influences of the process parameters and the mechanism were discussed in detail.

2 Experiment

2.1 Preparation of TaTiN coatings on Ti substrates

TA1 titanium substrates (15 mm×15 mm×1 mm) were ground with 400# – 5000# SiC sandpaper, then mirror-polished, and finally ultrasonically cleaned in anhydrous ethanol for 30 min. TaTiN coatings were deposited on polished Ti substrates by co-sputtering Ta and Ti targets (purity of 99.99% and Φ=50 mm) under nitrogen and argon sputtering gas. The Ta target was powered by a radio-frequency source at 100 W, and the Ti target was powered by a direct current source at 80 W. Following chamber evacuation to a base pressure (6.8×10⁻⁴ Pa), a 15-min pre-sputtering process was conducted before deposition to eliminate surface impurities. Table 1 summarizes the detailed coating fabrication parameters.

2.2 Characterization of TaTiN coatings

Surface and cross-sectional microstructures were characterized by a field-emission scanning electron microscope (SEM, JSM-7001F). We determined the mean grain size from statistically significant grain populations in SEM micrographs using ImageJ and averaged three measurements. We characterized

the coating phases by grazing incidence X-ray diffraction (XRD; Empyrean; Cu Kα radiation at 40 kV and 40 mA; incident angle of 0.6°; scan rate of 4°/min). We determined chemical bonding states by X-ray photoelectron spectroscopy (XPS, Thermo Fisher) using monochromatic Al Kα radiation. All spectra were energy-referenced to the C 1s peak at 284.8 eV. The surface roughness of the coatings was characterized using a confocal laser scanning microscope (Olympus).

2.3 Electrochemical tests

We assessed the corrosion resistance of TaTiN coatings using potentiodynamic and potentiostatic polarization tests and electrochemical impedance spectroscopy (EIS) with a three-electrode system and an electrochemical workstation (CHI760E, Shanghai Chenhua). The tests were conducted in a heated acidic electrolyte (0.5 mol/L H₂SO₄+2 μL/L F⁻, 80 °C) to simulate electrolyzer operating conditions. All tests were performed after 30 min of open-circuit potential stabilization. The TaTiN coating, Hg/Hg₂Cl₂/KCl, and a Pt foil were used as the working electrode, reference electrode, and counter electrode, respectively. The potentiodynamic polarization curve was acquired from -0.8 V to 2.0 V vs. saturated calomel electrode (SCE). EIS was measured from 0.01 Hz to 100 kHz with an amplitude of 10 mV.

2.4 Measurement of ICR and hydrophobicity

To measure the ICR values, a constant-current series circuit consisting of copper electrodes, carbon paper (CP), and the sample was adopted. Two pieces of CP were placed between the test sample and the gold-plated copper electrode to simulate actual PEMWE conditions. The contact pressure was progressively increased from 0.1 MPa to 2.0 MPa in 0.1 MPa steps. The resistance values were recorded at each pressure level. The resistance (*R*₁) was measured between two copper electrodes and the sample. Then, the resistance of the CP alone was recorded as *R*₂. The ICR can be evaluated as:

$$ICR = \frac{R_1 - R_2}{2}$$

(1)

The hydrophobicity of the TaTiN coating was evaluated by water contact angle using a meter (DSA30-KRÜSS) with deionized water droplets (5 μL). We took three measurements per sample and averaged the results.

3 Results and Discussion

3.1 Influence of N₂/Ar ratio on microstructure and properties

Fig. 1a₁–1a₄ and Fig. 1b₁–1b₄ show the surface and cross-sectional morphologies of TaTiN coatings. TaTiN coatings exhibit a uniform surface morphology with fine grains and no defects. Moreover, the average grain size increases with the N₂/Ar ratio (Fig. 1c and Fig.S1). Specifically, the coating deposited at a N₂/Ar ratio of 3:30 shows discontinuous morphology, with visible intergranular gaps, while the coatings deposited at the higher N₂/Ar ratios (4:30 and 5:30) exhibit a smoother and more continuous surface. However, as the N₂/Ar ratio increases further to 6:30, agglomeration and pores emerge (Fig. 1a₄). As shown in Fig. 1d, as the N₂/Ar ratio increases, the thickness of

Table 1 Deposition parameters of TaTiN coatings

Sample	Working pressure/Pa	N ₂ :Ar flow ratio	Temperature/°C	Time/h
N-3:30	0.90	3:30	200	1.5
N-4:30	0.90	4:30	200	1.5
N-5:30	0.90	5:30	200	1.5
N-6:30	0.90	6:30	200	1.5
P-0.83	0.83	5:30	200	1.5
P-0.90*	0.90	5:30	200	1.5
P-0.97	0.97	5:30	200	1.5
P-1.04	1.04	5:30	200	1.5
T-200**	0.97	5:30	200	1.5
T-300	0.97	5:30	300	1.5
T-400	0.97	5:30	400	1.5

Note: N-5:30 and P-0.90 refer to the same coating; P-0.97 and T-200 refer to the same coating.

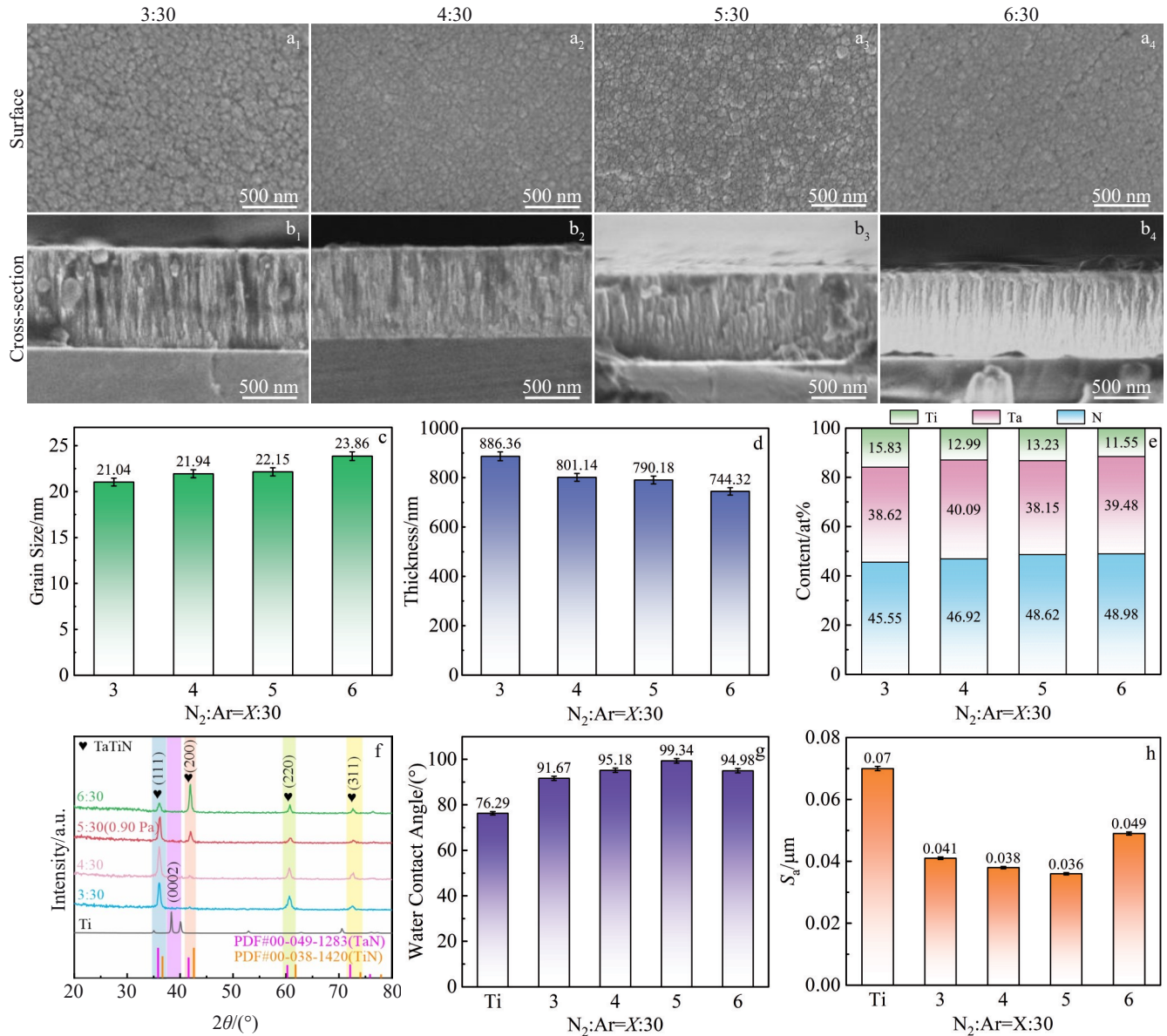


Fig.1 Surface (a_1 – a_4) and cross-sectional (b_1 – b_4) microstructures, grain size (c), thickness (d), element contents (e), XRD patterns (f), water contact angle (g), and surface roughness (h) of TaTiN coatings as a function of N_2/Ar ratios

TaTiN coatings decreases from about 886 nm to 744 nm, attributed to enhanced scattering of sputtered target atoms by the increased nitrogen content in the gas phase^[22–23]. Fig. 1e and Fig. S2 show the atomic contents in TaTiN coatings. The metal-to-nitrogen ratio ranges from approximately 54:46 to 51:49. Fig. 1f shows XRD patterns of the TaTiN coatings. Face-centered cubic (fcc) structure can be observed. The peaks at 36.15° , 41.85° , and 60.75° are assigned to the (111), (200), and (220) planes of the solid solution of δ -TaN (fcc, $a=0.4339$ nm) and δ -TiN (fcc, $a=0.241$ nm). Preferred (111) orientation is evidenced on the TaTiN coatings deposited at N_2/Ar ratios of 3:30, 4:30, and 5:30. However, as the N_2/Ar ratio increases to 6:30, the preferred orientation changes to (200) due to the competition between surface energy and strain energy^[24–26]. As shown in Fig. 1g and Fig. S3, TaTiN coatings deposited at N_2/Ar ratios of 3:30, 4:30, 5:30, and 6:30 have water contact angles of 91.67° , 95.18° , 99.34° , and 94.98° , respectively,

which are significantly higher than those of Ti substrates (76.29°). The TaTiN coating deposited at N_2/Ar of 5:30 has the largest water contact angle, showing characteristic of hydrophobicity^[27]. Fig. 1h and Fig. S4 present the surface roughness (S_a) of TaTiN coatings. The S_a of TaTiN coatings ranges from 0.036 μm to 0.049 μm , and the coating deposited at N_2/Ar of 5:30 has the lowest roughness, thereby enhancing both conductivity and corrosion resistance^[9,24]. Fig. S5 displays XPS spectra of TaTiN coatings. In the Ta 4f XPS spectra, the peaks at 23.8 and 25.7 eV correspond to the Ta 4f_{7/2} and Ta 4f_{5/2} orbitals^[28], respectively, which can be deconvoluted into doublets attributed to Ta-N bonds (23.7 and 25.5 eV) and Ta-O bonds (25.9 and 27.8 eV)^[23]. Similarly, the Ti 2p spectra in Fig. S5c are deconvoluted into three spin-orbit doublets, assigned to Ti-N (454.8 and 460.5 eV), Ti-N-O (456.4 and 462.1 eV), and Ti-O (458.2 and 463.7 eV) species.

Fig. 2a–2c present the potentiodynamic polarisation (PDP)

curves, corrosion current density I_{corr} , and corrosion potential U_{corr} of the TaTiN coatings on Ti substrates. Compared with Ti substrates, TaTiN coatings show significantly enhanced corrosion resistance, with a higher U_{corr} and a lower I_{corr} . The TaTiN coating deposited at N_2/Ar ratio of 5:30 presents the most positive U_{corr} (-0.508 V vs. SCE), the lowest I_{corr} ($1.41 \mu\text{A} \cdot \text{cm}^{-2}$), and the highest polarization resistance ($R_p = 3.03 \times 10^4 \Omega \cdot \text{cm}^2$) (Fig. 2d), indicating excellent corrosion resistance. Fig. 2e–2f show EIS results of TaTiN coatings. The Nyquist plot exhibits characteristic capacitive semicircles (Fig. 2e), which are fitted with an equivalent electrical circuit (EEC, inset) model consisting of solution resistance (R_s), charge-transfer resistance (R_{ct}), and a constant phase element (CPE) that models the non-ideal double-layer capacitance. CPE impedance is expressed as^[9]:

$$Z_{\text{CPE}} = Y_0^{-1} (j\omega)^{-n} \quad (2)$$

where Y_0 quantifies the admittance coefficient of the CPE, j is the imaginary unit, ω is the angular frequency, and n is the CPE index, a dimensionless parameter typically between 0 and 1. If n is 1 or 0, the CPE is equivalent to a capacitor or a

resistor, respectively. A larger n is typically associated with lower coating porosity, higher densification, and better protection^[29]. Table S1 lists the fitted parameters. The TaTiN coating deposited at a N_2/Ar ratio of 5:30 exhibits the largest capacitive arc semicircle in the Nyquist plot (Fig. 2e), indicating the highest R_{ct} . Furthermore, the Bode phase plot (Fig. 2f) shows the highest phase angle and the largest impedance modulus ($|Z|$) at low frequencies, suggesting the best stability. Fig. 2g shows the ICR values of TaTiN coatings at compaction forces from 0.1 MPa to 2.0 MPa, and Fig. 2h plots the ICR at 1.5 MPa, which decreases from about $19.58 \text{ m}\Omega \cdot \text{cm}^2$ to $15.19 \text{ m}\Omega \cdot \text{cm}^2$ as the N_2/Ar ratio increases from 3:30 to 5:30, and is much lower than that of the Ti substrate (about $39.49 \text{ m}\Omega \cdot \text{cm}^2$). However, the ICR of the TaTiN coating deposited at a N_2/Ar ratio of 6:30 increases significantly to about $47.48 \text{ m}\Omega \cdot \text{cm}^2$ due to the rougher surface and defects (Fig. S4).

3.2 Influence of working pressure on microstructure and properties

The influence of working pressure in the range of 0.83–

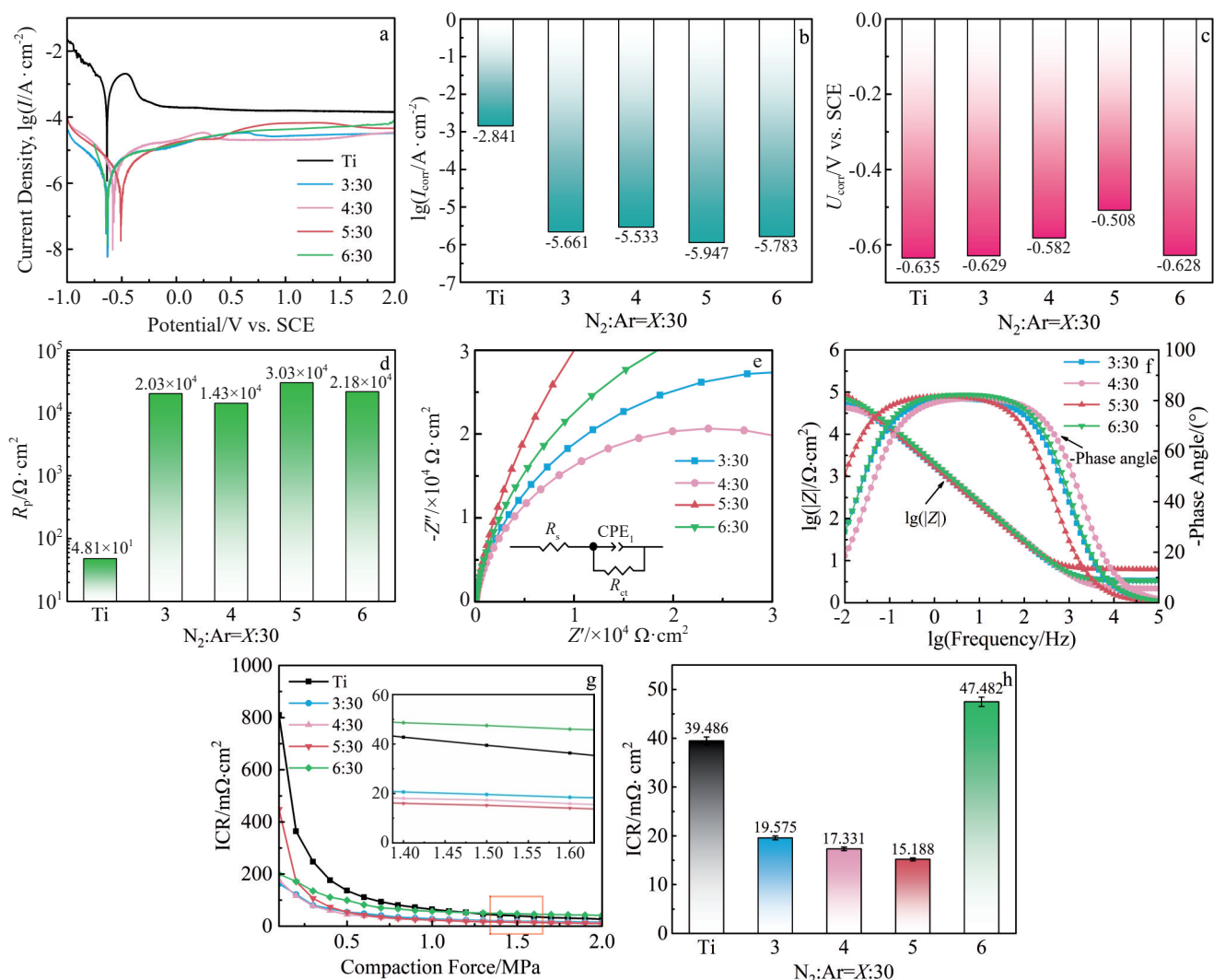


Fig.2 Performance metrics of TaTiN coatings deposited at varying N_2/Ar flow ratios: (a) PDP curves, (b) I_{corr} , (c) U_{corr} , (d) polarization resistance, (e) Nyquist plot, (f) Bode plot, (g) ICR at 0.1–2.0 MPa, and (h) ICR at 1.5 MPa

1.04 Pa is also investigated. The surface/cross-sectional SEM images are displayed in Fig. 3a–3c. TaTiN coatings exhibit uniform surfaces with a grain size of 19–23 nm (Fig. 3d and Fig. S6), resulting from competition between Ti-doping-induced grain refinement and large working-pressure-induced grain coarsening^[30–31]. The thickness of TaTiN coatings increases gradually from about 740 nm to 906 nm with increasing working pressure (Fig. 3e), due to the enhanced sputtering efficiency^[32], consistent with the increased sputtering yields of Ta and Ti (Fig. 3f and Fig. S7). Accordingly, the atomic percentage of Ti increases from 11.47at% to 18.38at%, whereas that of N decreases from 49.26at% to 43.63at%. Fig. 3g presents XRD patterns of TaTiN coatings. The peaks at 36.15°, 41.85°, and 60.75° correspond to (111), (200), and (220) planes of TaTiN coatings, respectively. As the working pressure increases, the preferred orientation changes from the (200) plane to the (111) plane. The water contact angle on the TaTiN coating deposited at the working pressure of 0.97 Pa is the largest one (101.87°), indicating excellent hydrophobicity (Fig. 3h)^[33]. Essentially, increasing Ti content not only refines grains and

enhances densification but also modifies surface chemical bonding states, thereby increasing the water contact angle. As the working pressure increases to 1.04 Pa, the surface porosity of the coatings increases, and hydrophobicity decreases (Fig. S8). Fig. 3i and Fig. S9 show the roughness and morphologies of TaTiN coatings. A parallel, point-like morphology is evident, owing to scratches on the Ti substrates, and the surface roughness decreases initially and then increases with increasing working pressure. The coating deposited at the working pressure of 0.97 Pa has the minimum surface roughness of 0.035 μm . XPS spectra indicate that the coatings are primarily composed of metal nitrides and oxides (Fig. S10). The spin-orbit doublets of Ta 4f (23.8 and 25.7 eV) and Ti 2p (455.4 and 460.8 eV) are assigned to δ -TaN and δ -TiN, respectively. The O 1s peaks (529.8 and 531.9 eV) are attributed to metal oxides and hydroxyl groups, respectively (Fig. S10b). In the N 1s spectra (Fig. S10e), the peak at 396.2 eV is assigned to N-metal bonds, while the peak at 402.6 eV is assigned to the Ta 4p_{3/2} orbit^[34].

Fig. 4a–4c show the PDP curves, I_{corr} , and U_{corr} of TaTiN coatings on Ti substrates prepared at different working pressures,

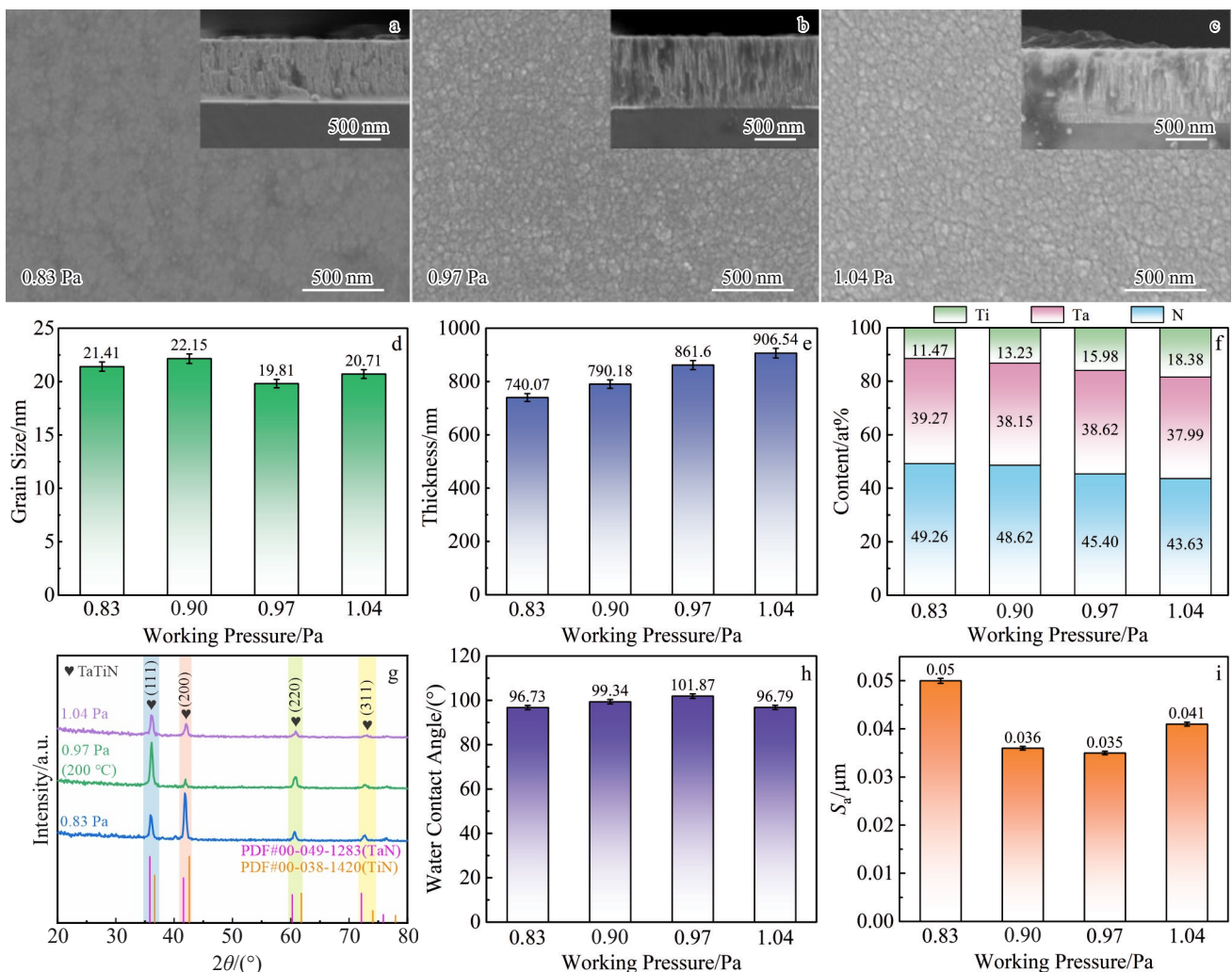


Fig.3 Characterization of TaTiN coatings prepared at different working pressures: (a–c) surface and cross-sectional SEM images, (d) grain size, (e) thickness, (f) element contents, (g) XRD patterns, (h) water contact angle, and (i) surface roughness

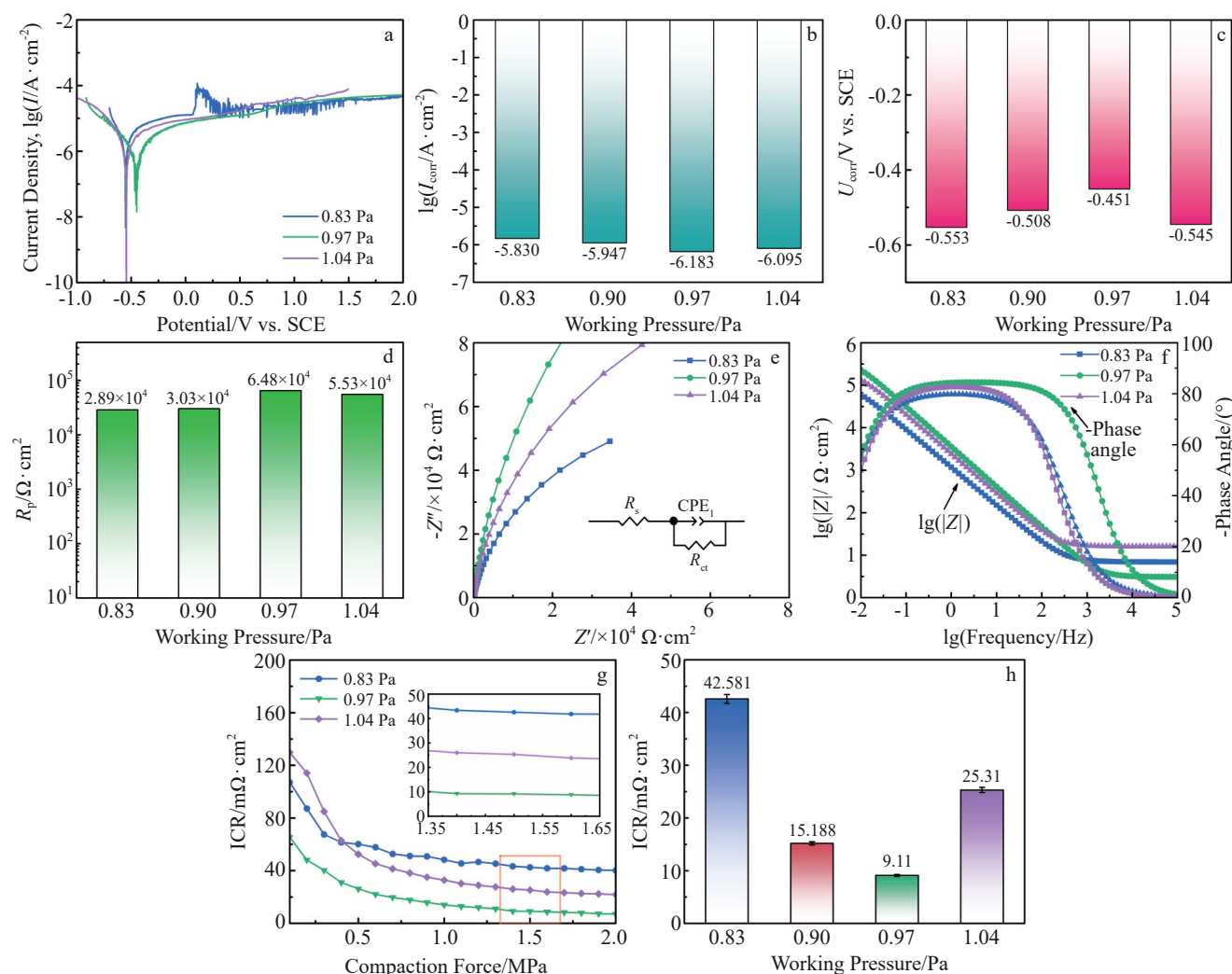


Fig.4 PDP curves (a), I_{corr} (b), U_{corr} (c), R_p (d), Nyquist plot (e), Bode plot (f), and ICR (g–h) of TaTiN coatings prepared at different working pressures

respectively. The TaTiN coating deposited at 0.97 Pa presents the most positive U_{corr} (-0.451 V vs. SCE), the lowest I_{corr} ($6.56 \times 10^{-7} \text{ A} \cdot \text{cm}^{-2}$), the highest R_p ($6.48 \times 10^4 \Omega \cdot \text{cm}^2$, Fig. 4d), and the highest R_{ct} ($4.67 \times 10^5 \Omega \cdot \text{cm}^2$, Fig. 4e, Table S2), indicating the strongest corrosion resistance. Fig. 4f displays the Bode plot of this coating. A higher and broader phase-angle plateau is evident in the medium-to-low frequency range, suggesting that the dense, stable protective layer can effectively inhibit the penetration of corrosive ions toward the underlying metal substrate^[35]. As shown in Fig. 4g–4h, the ICR values of TaTiN coatings decrease initially and then increase with increasing working pressure. The TaTiN coating prepared at 0.97 Pa shows the minimum ICR value of $9.11 \text{ m}\Omega \cdot \text{cm}^2$ at 1.5 MPa, owing to its smooth surface (low roughness).

3.3 Influence of substrate temperature on microstructure and properties

The effects of substrate temperature ($200 - 400^\circ \text{C}$) are studied. Fig. 5a–5b show the resulting microstructural changes. As the substrate temperature increases from 200°C to 400°C , the grain size in the TaTiN coating increases (Fig. 5c and Fig. S11), accompanied by significant agglomeration at 400°C

(Fig. 5b)^[36]. The coating thickness ranges from 843 nm to 881 nm. As shown in Fig. 5d–5f and Fig. S12, scratches and pores are visible on the coatings, and these surface features are particularly pronounced on the coating prepared at 400°C . Surface roughness increases from $0.035 \mu\text{m}$ to $0.056 \mu\text{m}$. As shown in XRD patterns (Fig. 5g), the peaks at 36.15° , 41.85° , and 60.75° can be ascribed to TaTiN coatings, while the peak at 38.55° originates from Ta_2N . The preferred orientation changes from (111) to (200) as the substrate temperature increases from 200°C to 300°C . However, the coatings deposited at 400°C show no preferential orientation. The atomic composition in the coatings changes significantly with deposition temperature (Fig. 5h and Fig. S13). The Ta content decreases first and then increases to 43.67at%, while the Ti content increases steadily from 15.98at% to 26.37at%. The N content declines from 45.40at% to 29.95at%. This indicates the formation of a highly metal-rich, nitrogen-deficient coating, as well as the Ta_2N phase^[37–38]. The water contact angle of TaTiN coatings decreases from 101.87° to 96.03° with increasing substrate temperature (Fig. 5i and Fig. S14). XPS spectra (Fig. S15) indicate that the coating surface consists mainly of nitrides and oxides. XPS peaks at 454.8 (Ti $2p_{3/2}$)

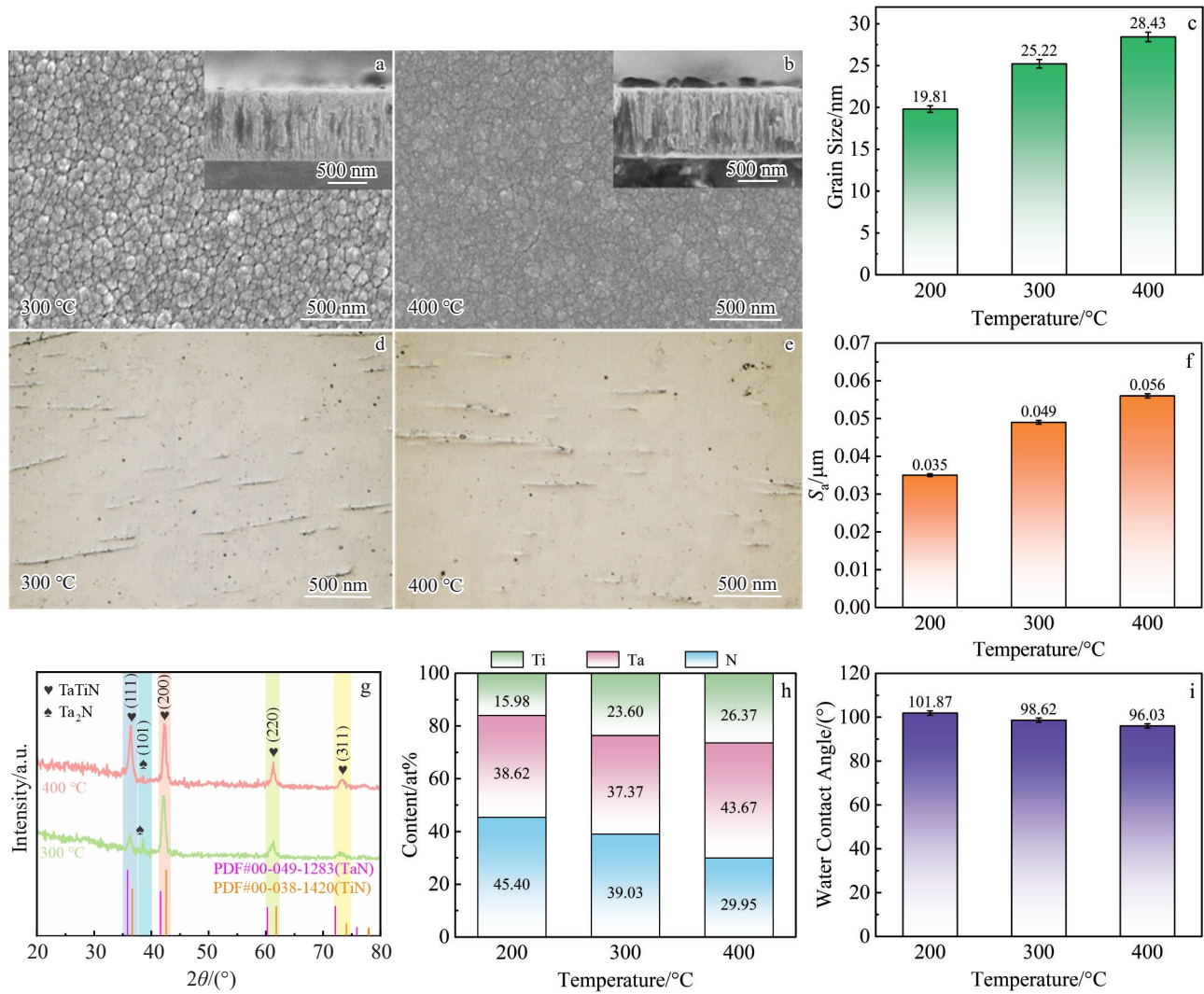


Fig.5 Influences of substrate temperature (300 and 400 °C) on TaTiN coatings: (a–b) surface and cross-sectional microstructures; (c) grain size; (d–e) laser confocal surface morphologies; (f) surface roughness; (g) XRD patterns; (h) element contents; (i) water contact angle

and 460.5 eV ($\text{Ti}2p_{1/2}$) are ascribed to Ti-N bonds. Notably, the intensity of Ti 2p peaks decreases with increasing substrate temperature, suggesting a reduced nitride content. XPS peak at 23.1 eV belongs to Ta_2N .

Fig. 6a–6c show the PDP curves, I_{corr} , and U_{corr} values of TaTiN coatings deposited at different temperatures. The TaTiN coatings deposited at 300 and 400 °C show inferior corrosion resistance compared with the coating deposited at 200 °C. Moreover, coatings deposited at 300 and 400 °C show lower R_p values and degraded corrosion resistance (Fig. 6d). Fig. 6e–6f and Table S3 show EIS spectra of the TaTiN coatings. The TaTiN coating deposited at 300 °C shows better corrosion performance than that deposited at 400 °C, as indicated by the larger R_{ct} and higher phase angle. In contrast, the TaTiN coating deposited at 400 °C shows a narrower phase-angle plateau and lower $|Z|$, indicating degraded corrosion protection. This mainly stems from increased surface roughness, porosity, and grain coarsening in the coatings. The ICR at 1.5 MPa increases with substrate temperature (Fig. 6g–6h), partly due to increased porosity and structural defects at

elevated temperatures (Fig. 5d–5e).

3.4 Stability and corrosion behavior of TaTiN coating during polarization

The stability of the optimized TaTiN coating deposited at a N_2/Ar ratio of 5:30, a working pressure of 0.97 Pa, and 200 °C is evaluated through potentiostatic polarization for 3 h. Fig. 7a presents the current density evolution of the Ti substrate and the TaTiN coating during 3 h of polarization at 2.0 V. The Ti substrate is unstable, as shown by substantial variations in current density. In contrast, the TaTiN coating maintains a stable corrosion current density of about $30 \mu\text{A}\cdot\text{cm}^{-2}$, demonstrating significantly high corrosion resistance and stability. Fig. 7b–7c show EIS spectra of the TaTiN coating after 3 h of potentiostatic polarization, in which an EEC model with dual time constants is shown in Fig. 7b. R_s is the solution resistance, CPE_1 is the constant phase element for the solution/coating double-layer capacitance, R_c denotes the resistance of the passive layer, CPE_2 corresponds to the coating/metal interface double-layer capacitance, and R_{ct} is the charge transfer resistance^[39]. After 3 h of potentiostatic

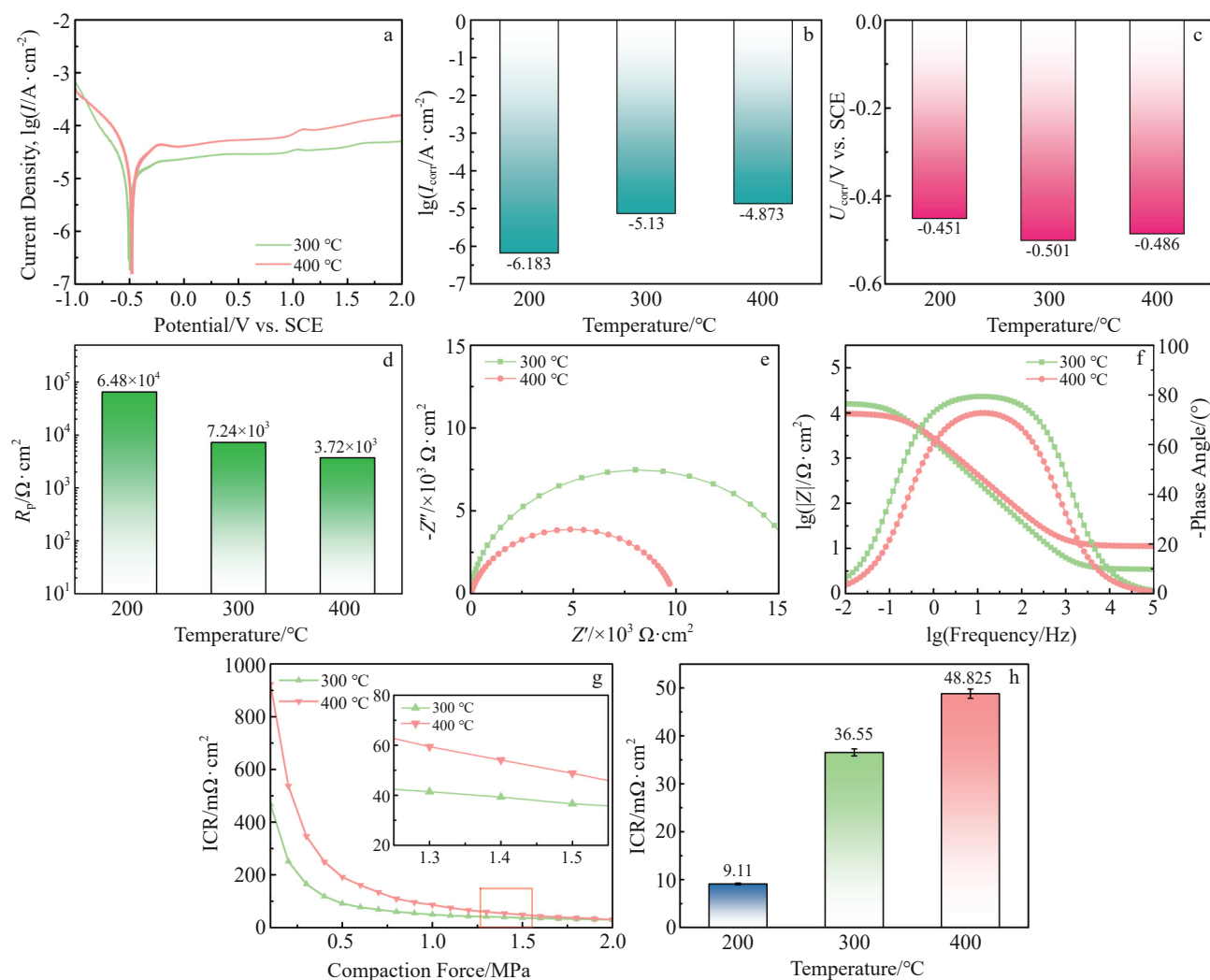
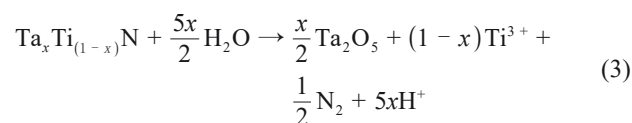


Fig.6 PDP curves (a), I_{corr} (b), U_{corr} (c), R_p (d), Nyquist plot (e), Bode plot (f), and ICR (g–h) of TaTiN coatings prepared at different temperatures

polarization, the R_{ct} value of the TaTiN coating is $4.27 \times 10^5 \Omega \cdot \text{cm}^2$, showing a reduction of only 8.57% compared with the value before potentiostatic polarization (Table S4). Furthermore, the coating maintains a large angle of -76° in the medium-frequency range, along with a large $|Z|$ and phase angle at 0.01 Hz. The values of n_1 and n_2 corresponding to CPE_1 and CPE_2 are 0.74 and 0.95, respectively (Table S4). These results indicate that corrosion is primarily confined to the surface, while the coating interior remains uniform and dense, exhibiting near-ideal capacitive behavior. The results confirm that the TaTiN coating maintains excellent protective properties even after 3 h of potentiostatic polarization. However, the ICR value is increased to $105.65 \text{ m}\Omega \cdot \text{cm}^2$ due to the formation of a stable passive layer on the TaTiN coatings, but remains lower than that of corroded Ti ($375.86 \text{ m}\Omega \cdot \text{cm}^2$) (Fig. 7d – 7e). The chemical states of the polarized TaTiN coatings are characterized by XPS (Fig. 7f–7i). In the Ti 2p spectra (Fig. 7f), the Ti 2p peaks (458.5 and 464.6 eV) indicate Ti-O bonding states, but no Ti-N signal is detected. In the Ta 4f spectra (Fig. 7g), the Ta-O signal dominates, with a weak Ta-N peak corresponding to 85.63% Ta-O bonds and

14.37% Ta-N bonds. In the O 1s spectra (Fig. 7i), a substantial amount of surface hydroxyl (OH) groups is evidenced, due to water adsorption^[40].

The calculated U -pH diagram shows that TaTiN corrosion behavior evolves with applied potential, delineating three distinct regions (Fig. 8a)^[41]. At low potentials, TaTiN resides in an immunity region, and the coating remains in its solid solution phase without spontaneous corrosion or dissolution. In acidic environments and in the voltage range from -0.4 V to 0.28 V vs. standard hydrogen electrode (SHE), selective dissolution occurs, releasing soluble Ti^{3+} ions and forming a Ta_2O_5 passivation layer. Moreover, at higher potentials, the system enters a well-defined passivation region, in which the TaTiN coating spontaneously oxidizes into a dense $\text{Ta}_2\text{O}_5/\text{TiO}_2$ composite layer that protects the coating from further degradation. The governing reactions are as follows:



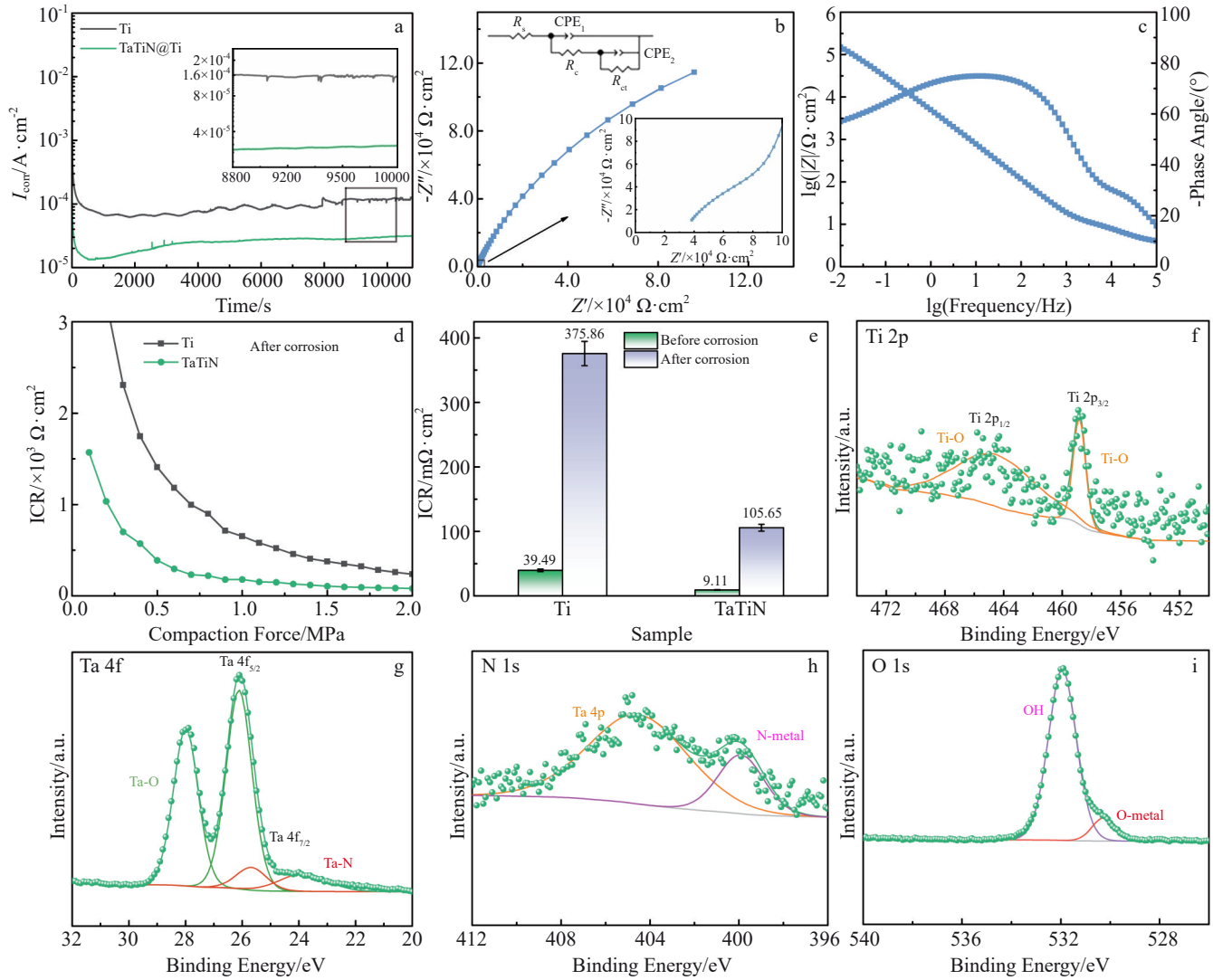
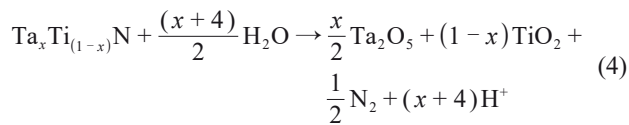


Fig.7 Current density of Ti substrate and TaTiN coatings during 3 h potentiostatic polarization test at 2 V ($N_2/Ar=5:30$, working pressure=0.97 Pa, and temperature=200 °C) (a); Nyquist plot (b) and Bode plot (c) after 3 h potentiostatic polarization; ICR for TaTiN coatings and Ti substrate at 0.1–2.0 MPa (d) and 1.5 MPa (e) before and after 3 h potentiostatic polarization; fitted XPS spectra of Ti (f), Ta (g), N (h), and O (i) of the TaTiN coating after 3 h potentiostatic polarization



SEM images of Ti and TaTiN before and after 3 h of potentiostatic polarization are displayed in Fig. 8b – 8e. Numerous pits and micropores appear on the Ti surface. Meanwhile, the TaTiN coating remains relatively smooth and intact, with no evidence of large-scale pitting or coating peeling after potentiostatic polarization. This direct morphological comparison confirms superior corrosion resistance.

3.5 Enhancement mechanism of electrical conductivity and corrosion resistance in TaTiN coatings

The TaTiN coating deposited at 200 °C with a N_2/Ar ratio of 5:30 and a working pressure of 0.97 Pa possesses a strong (111) preferred orientation, refined grains, and lower surface

roughness, and thus exhibits excellent corrosion resistance and high electrical conductivity. Furthermore, the TaTiN coating facilitates the formation of a denser, more continuous Ta_2O_5/TiO_2 mixed passivation layer during potentiostatic polarization, which inhibits the diffusion of aggressive species and consequently improves the corrosion performance of the coating system. Fig.9 illustrates the schematic of the corrosion protection mechanism of TaTiN coatings. Compared with the TaTiN (200)/Ti (0001) interface, the TaTiN (111)/Ti (0001) interface has a smaller lattice mismatch and stronger interfacial bonding, and the closely packed (111) plane favors a denser texture. Moreover, grain refinement can smooth the surface and facilitate the formation of a continuous, compact passive layer. Consequently, corrosion resistance improves. In fact, the lattice mismatch (δ) between the TaTiN coating and the Ti substrate is calculated as:

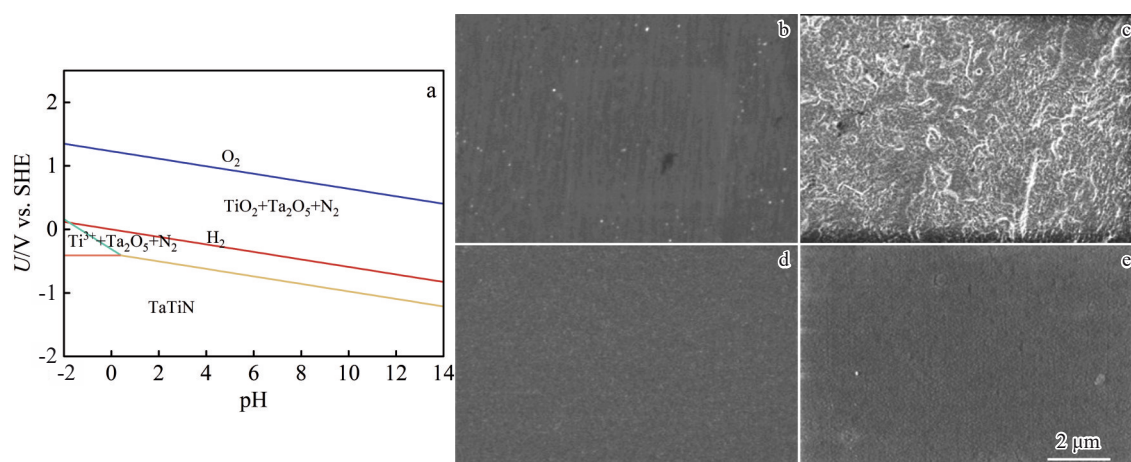


Fig.8 U - pH diagram of TaTiN (a); surface morphologies before and after 3 h of polarization at 2 V: (b-c) Ti and (d-e) TaTiN coating

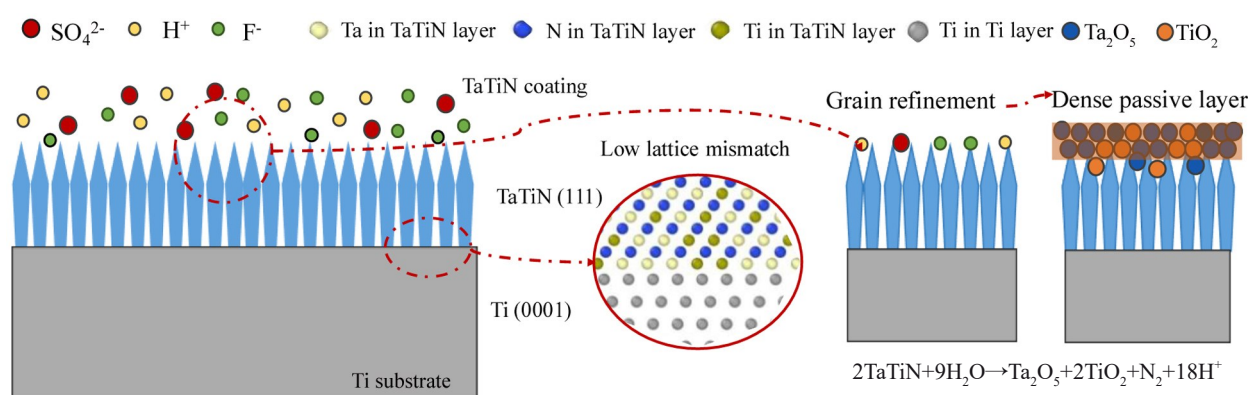


Fig.9 Corrosion protection mechanism schematic of TaTiN coatings

$$\delta = \frac{|a_1 - a_2|}{a_2} \quad (5)$$

in which a_1 and a_2 are the lattice constants of the coating and the substrate, respectively. As illustrated in Fig. 1f, the Ti substrate exhibits a preferred (0002) orientation, corresponding to the (0001) basal plane. Both the Ti (0001) and TaTiN (111) planes exhibit hexagonal symmetry, yielding an epitaxial relationship of $[11\bar{2}0]_{Ti} \parallel [1\bar{1}0]_{TaTiN}$ with a lattice mismatch of 2.73%, indicative of a semi-coherent interface^[42-43]. In contrast, Ti (0001) and TaTiN (200) planes exhibit a significantly higher lattice mismatch of 27.2%. The small lattice mismatch between TaTiN (111) and Ti (0001) can enhance interfacial bonding and thus electrical conductivity across the interface^[21,44-45], resulting in a lower ICR value.

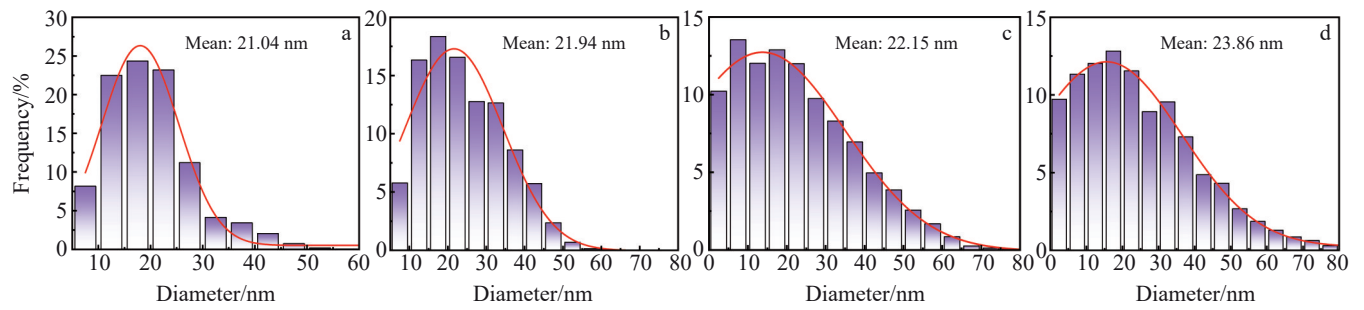
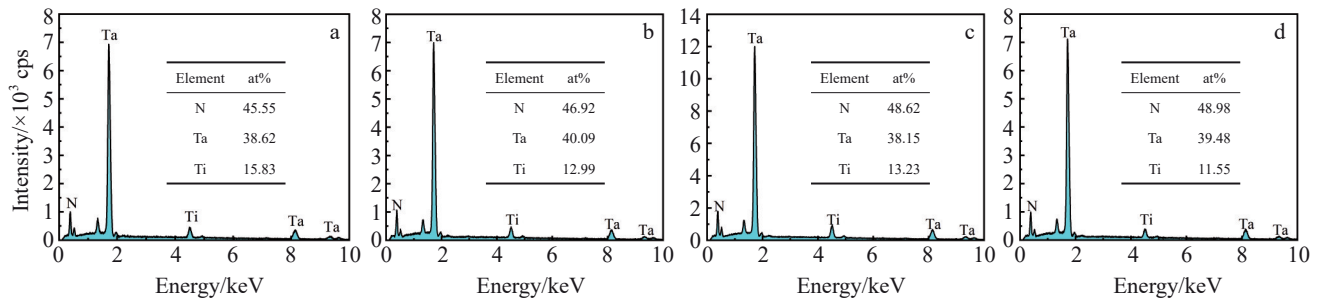
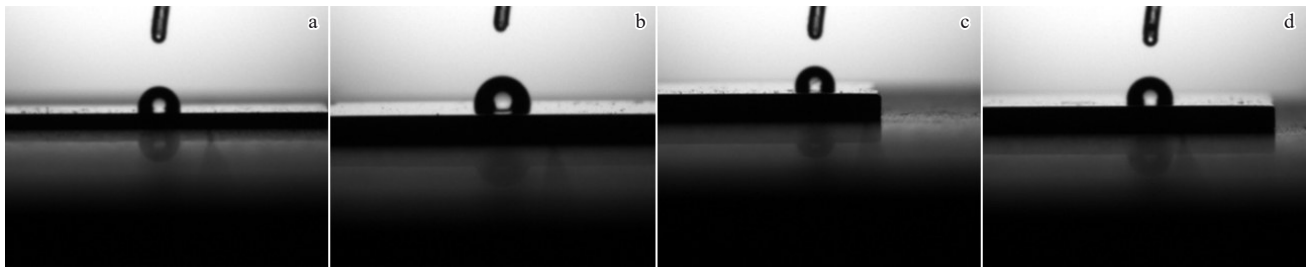
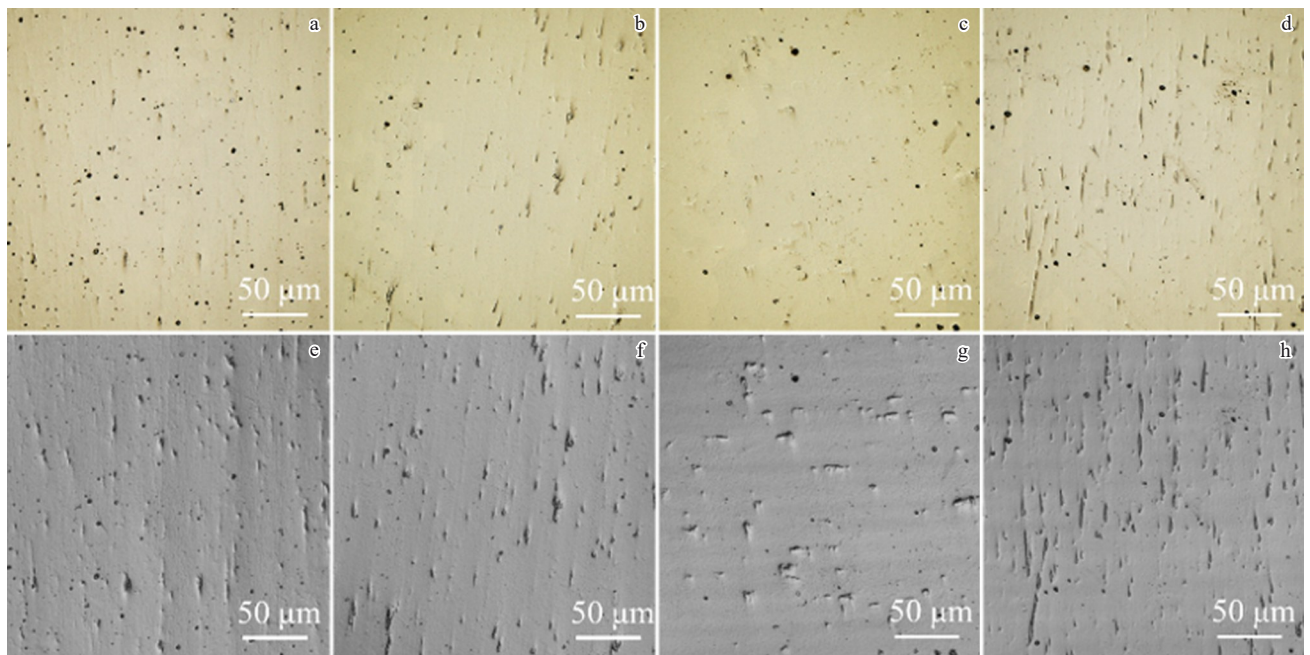
Commonly, a denser, more uniform passive layer forms on the closely packed (111) plane^[41]. Meanwhile, grain refinement can reduce surface porosity and promote dense coating formation. Together, these factors facilitate the growth of a continuous, dense passivation layer, thereby impeding further ingress of aggressive species^[46-50]. Conversely, intergranular voids in coarse columnar structures disrupt passive-layer uniformity, compromising its protective capability^[51].

4 Conclusions

1) TaTiN coatings are deposited on Ti bipolar plates via magnetron sputtering. Coatings deposited at 200 °C with a N_2/Ar ratio of 5:30 and a working pressure of 0.97 Pa have a (111) preferred orientation and fine grains, and exhibit the lowest I_{corr} ($6.56 \times 10^{-7} A \cdot cm^{-2}$) and ICR ($9.11 m\Omega \cdot cm^2$ at 1.5 MPa). On the one hand, the closely-packed (111) plane favors a dense texture; moreover, grain refinement can smooth the surface and facilitate the formation of a continuous, compact passive layer, improving corrosion resistance. In contrast, the small lattice mismatch between TaTiN (111) and Ti (0001) enhances interfacial bonding and, in turn, electrical conductivity across the interface.

2) The TaTiN coating exhibits a large water contact angle of 101.87° , indicating favorable hydrophobicity for water management in PEMWE. Remarkably, the TaTiN coating maintains a uniform, dense morphology and a minimal corrosion current density after 3 h of potentiostatic polarization. Therefore, the TaTiN coating offers an attractive, cost-effective strategy for protecting metallic BPs in PEMWE applications.

Supplement

Fig.S1 Grain size of the TaTiN coatings at different $N_2:Ar$ flow ratios: (a) 3:30, (b) 4:30, (c) 5:30, and (d) 6:30Fig.S2 EDS results of the TaTiN coatings at different $N_2:Ar$ flow ratios: (a) 3:30, (b) 4:30, (c) 5:30, and (d) 6:30Fig.S3 Water contact angles of the TaTiN coatings at different $N_2:Ar$ flow ratios: (a) 3:30; (b) 4:30; (c) 5:30; (d) 6:30Fig.S4 Laser confocal surface morphologies of the TaTiN coatings at different $N_2:Ar$ flow ratios: (a, e) 3:30; (b, f) 4:30; (c, g) 5:30; (d, h) 6:30

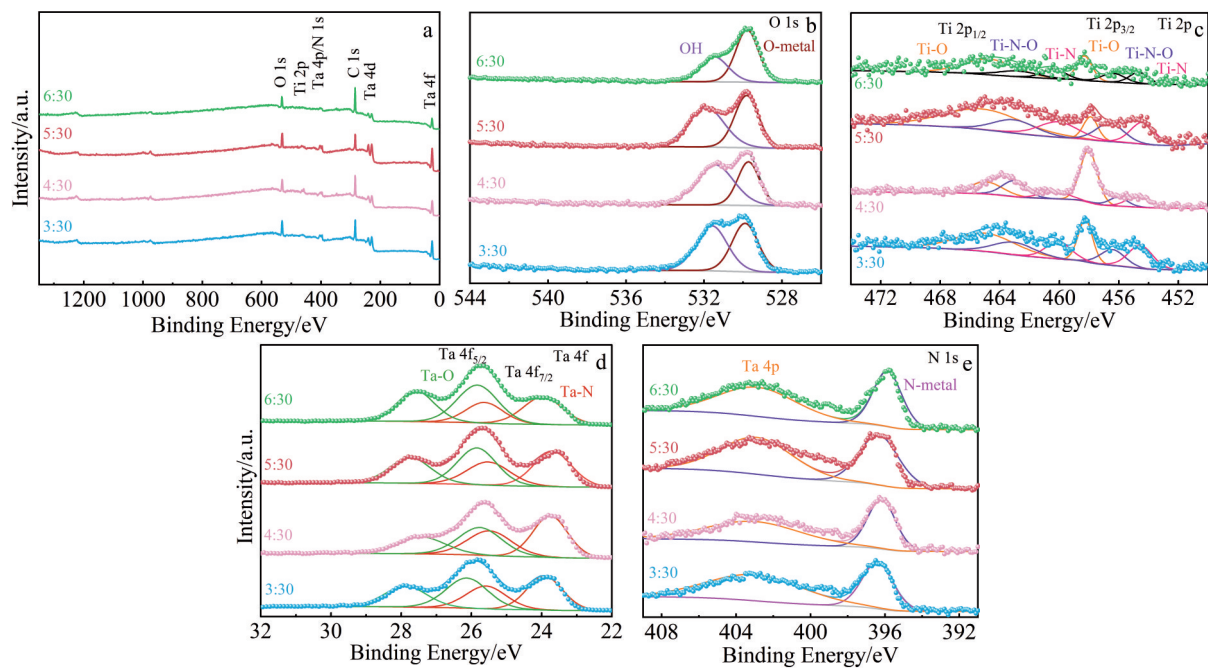


Fig.S5 XPS spectra of the TaTiN coatings prepared at different N₂/Ar flow ratios: (a) full XPS spectra, (b) O 1s, (c) Ti 2p, (d) Ta 4f, and (e) N 1s

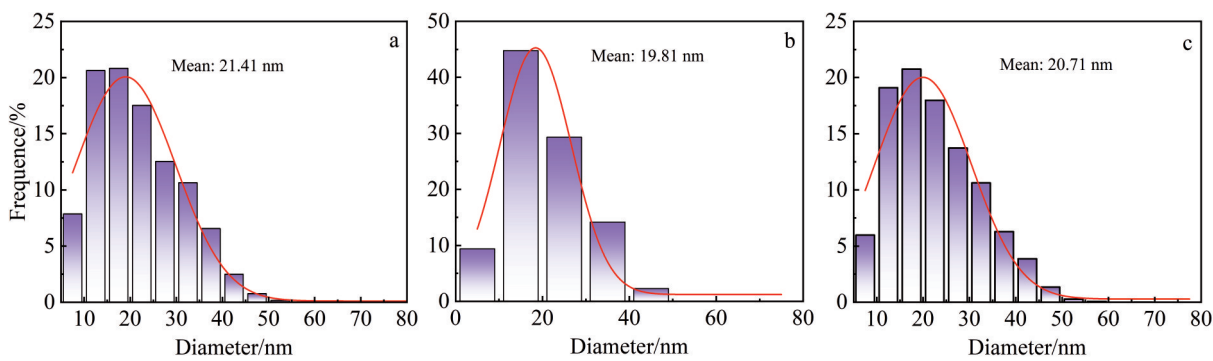


Fig.S6 Grain size of the TaTiN coatings at different working pressures: (a) 0.83 Pa; (b) 0.97 Pa; (c) 1.04 Pa

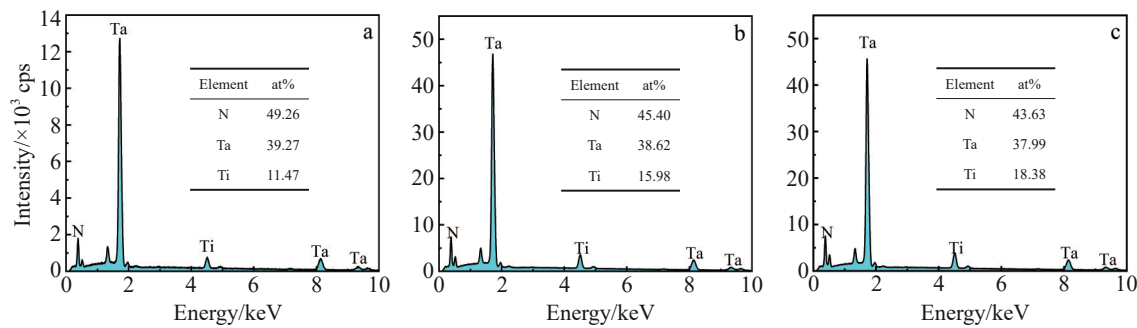


Fig.S7 EDS results of the TaTiN coatings at different working pressures: (a) 0.83 Pa; (b) 0.97 Pa; (c) 1.04 Pa

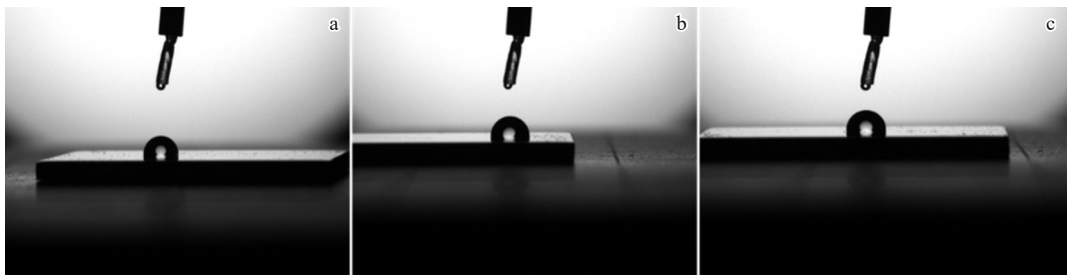


Fig.S8 Water contact angles of the TaTiN coatings at different working pressures: (a) 0.83 Pa; (b) 0.97 Pa; (c) 1.04 Pa

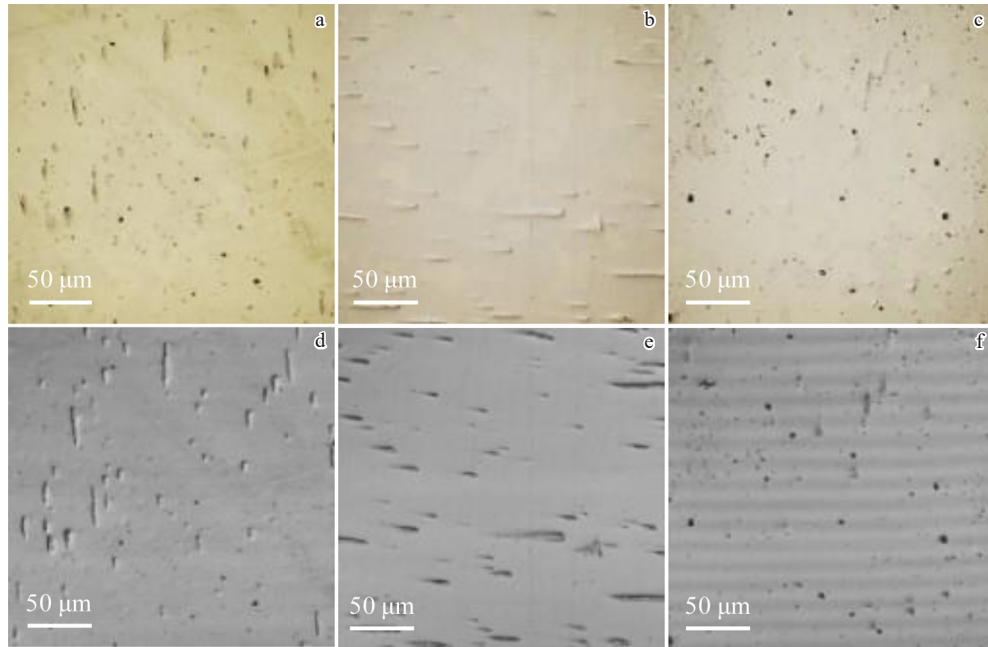


Fig.S9 Laser confocal surface morphologies of the TaTiN coatings at different working pressures: (a, d) 0.83 Pa; (b, e) 0.97 Pa; (c, f) 1.04 Pa

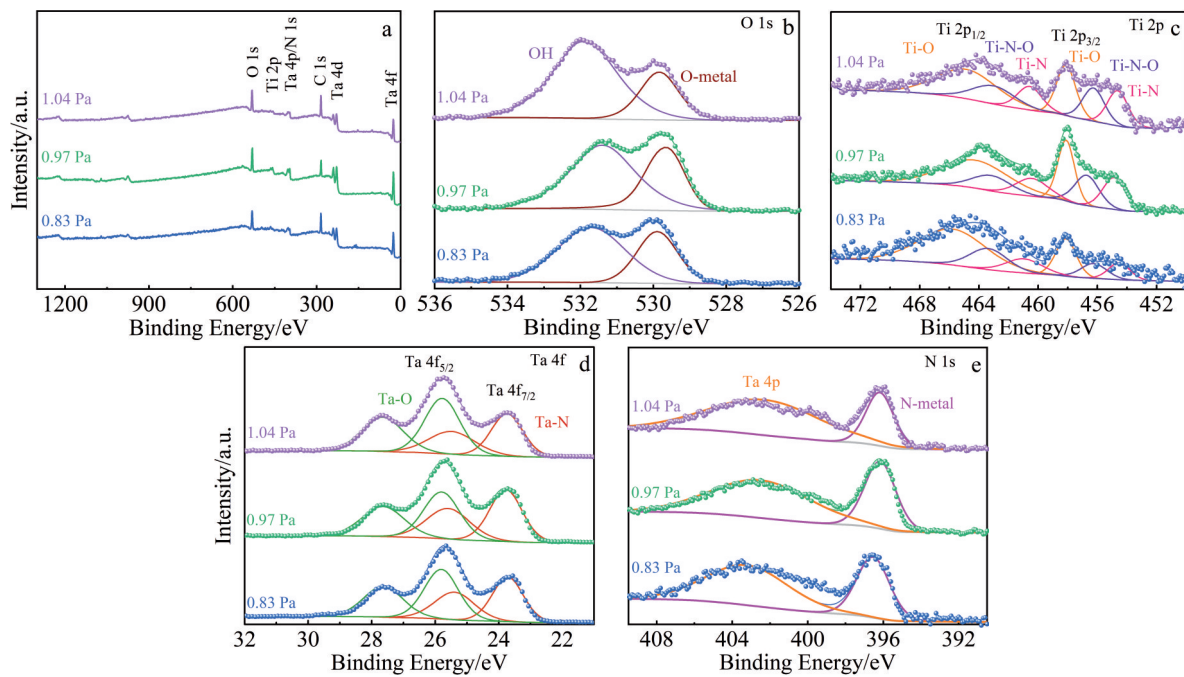


Fig.S10 XPS spectra of the TaTiN coatings prepared at different working pressures: (a) full XPS spectra, (b) O 1s, (c) Ti 2p, (d) Ta 4f, and (e) N 1s

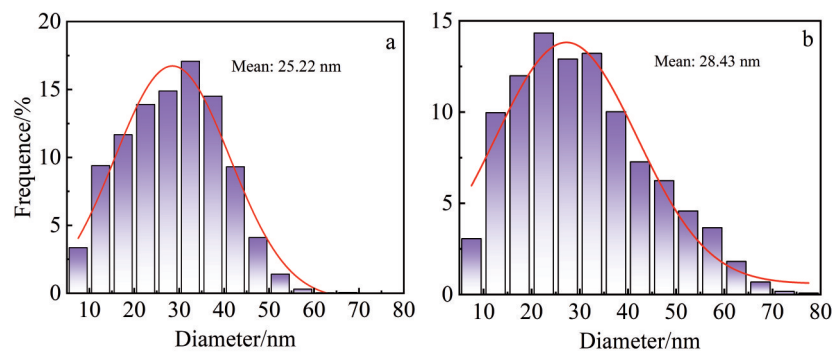


Fig.S11 Grain size of the TaTiN coatings at different deposition temperatures: (a) 300 °C and (b) 400 °C

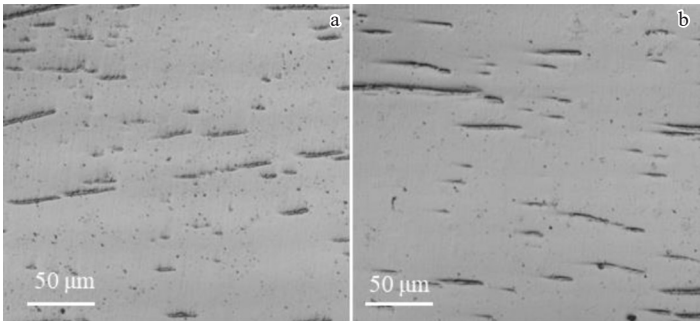


Fig.S12 Laser confocal surface morphologies of the TaTiN coatings at different deposition temperatures: (a) 300 °C and (b) 400 °C

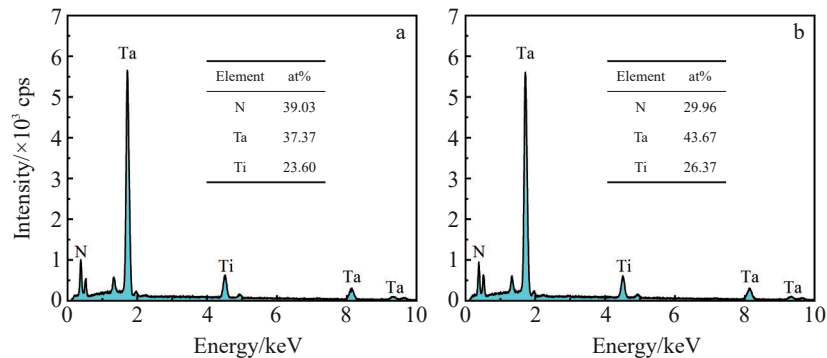


Fig.S13 EDS patterns of the TaTiN coatings at different deposition temperatures: (a) 300 °C and (b) 400 °C

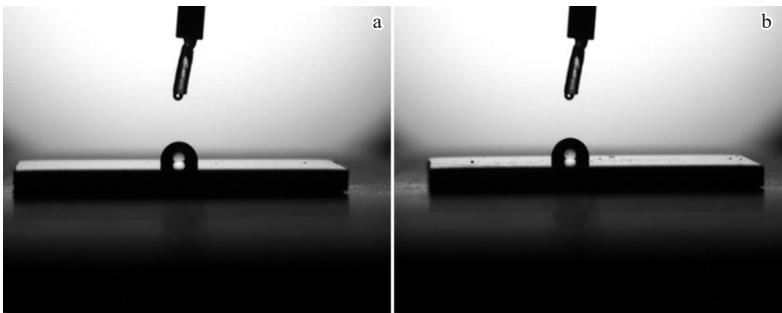


Fig.S14 Water contact angles of the TaTiN coatings at different deposition temperatures: (a) 300 °C and (b) 400 °C

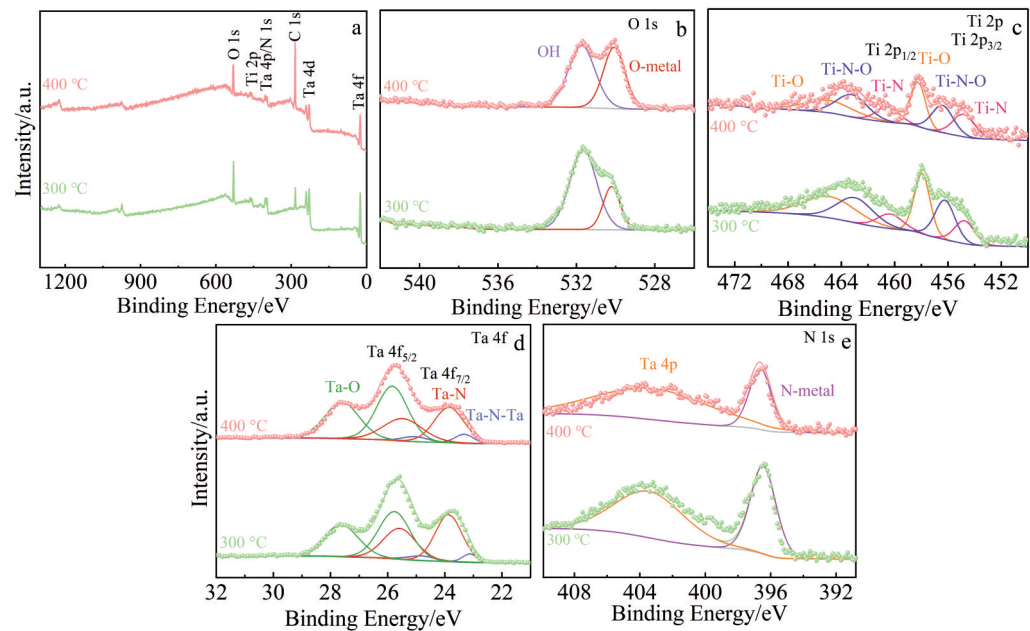


Fig.S15 XPS spectra of the TaTiN coatings prepared at different deposition temperatures: (a) full XPS spectra, (b) O 1s, (c) Ti 2p, (d) Ta 4f, and (e) N 1s

Table S1 EIS results of the TaTiN coatings deposited at different N₂:Ar flow ratios

N ₂ :Ar flow ratio	$R_s/\Omega\cdot\text{cm}^2$	$\text{CPE}_1/\Omega^{-1}\cdot\text{cm}^{-2}\cdot\text{s}^{-n}$	n_1	$R_{ct}/\Omega\cdot\text{cm}^2$
3:30	3.38	1.125×10^{-4}	0.90	6.39×10^4
4:30	2.18	9.230×10^{-5}	0.90	4.78×10^4
5:30	6.22	1.103×10^{-4}	0.92	1.68×10^5
6:30	3.37	8.714×10^{-5}	0.91	8.41×10^4

Table S2 EIS results of the TaTiN coatings deposited at different working pressures

Pressure/Pa	$R_s/\Omega\cdot\text{cm}^2$	$\text{CPE}_1/\Omega^{-1}\cdot\text{cm}^{-2}\cdot\text{s}^{-n}$	n_1	$R_{ct}/\times 10^5 \Omega\cdot\text{cm}^2$
0.83	6.92	1.645×10^{-4}	0.90	1.36
0.97	3.14	6.191×10^{-5}	0.95	4.67
1.04	15.60	7.545×10^{-5}	0.93	2.40

Table S3 EIS results of the TaTiN coatings deposited at different temperatures

Temperature/°C	$R_s/\Omega\cdot\text{cm}^2$	$\text{CPE}_1/\times 10^{-5} \Omega^{-1}\cdot\text{cm}^{-2}\cdot\text{s}^{-n}$	n_1	$R_{ct}/\Omega\cdot\text{cm}^2$
300	3.455	8.081	0.90	1.633×10^4
400	11.28	6.818	0.85	9.859×10^3

Table S4 EIS results of the TaTiN coating after potentiostatic polarizations

$R_s/\Omega\cdot\text{cm}^2$	$\text{CPE}_1/\Omega^{-1}\cdot\text{cm}^{-2}\cdot\text{s}^{-n}$	n_1	R_c	$\text{CPE}_2/\times 10^{-6} \Omega^{-1}\cdot\text{cm}^{-2}\cdot\text{s}^{-n}$	n_2	$R_{ct}/\Omega\cdot\text{cm}^2$
3.262	3.791×10^{-5}	0.74	13.91	9.341	0.95	4.27×10^5

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钛双极板 TaTiN 涂层耐腐蚀性与导电性的协同增强

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摘 要: 质子交换膜水电解 (PEMWE) 的大规模应用迫切需要双极板具有强耐腐蚀性和高导电性的涂层。通过磁控溅射制备了高性能 TaTiN 涂层。结果表明, 具有(111)晶面择优取向和晶粒细化的 TaTiN 涂层表现出更高的耐腐蚀性和更好的导电性。一方面, 密排(111)晶面有利于形成致密结构, 同时晶粒细化可使表面平整, 促进连续致密钝化层的形成, 从而提升耐腐蚀性能。另一方面, TaTiN (111)晶面与 Ti (0001)基底之间较小的晶格失配增强了界面结合, 进而改善了界面导电性, 降低界面接触电阻 (ICR)。优化后的 TaTiN 涂层腐蚀电流密度低至 $6.56 \times 10^{-7} \text{ A} \cdot \text{cm}^{-2}$, ICR 值仅为 $9.11 \text{ m}\Omega \cdot \text{cm}^2$, 表明 TaTiN 涂层在保护 PEMWE 系统中金属双极板方面具有巨大潜力。

关键词: TaTiN 涂层; 耐腐蚀性; 导电性; 双极板; 质子交换膜电解槽

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