

# Efficient Preparation of Nanoparticles Reinforced Nickel-based Composite Coating with Highly Preferred (220) Orientation

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## Abstract

This paper reports a phenomenon that the grain orientation gradually evolves to (220) with the deposition current density increasing when preparing nanoparticles reinforced nickel-based composite coatings by jet electrodeposition (JED). During the preparation of Ni-SiC composite coatings, the deposition current density increases from 180 A/dm<sup>2</sup> to 220 A/dm<sup>2</sup>, and TC(220) gradually increases from 41.4% to 97.7% correspondingly.

The Ni-SiC composite coating with highly preferred (220) orientation has superior corrosion resistance and adhesion force. With the increase of TC(220), the surface roughness is reduced from Ra1.210 μm to Ra0.119 μm, the self-corrosion potential increases from -0.747 V to -0.477 V, the corrosion current density decreases from 54.52 μA·cm<sup>2</sup> to 2.76 μA·cm<sup>2</sup>, the diameter of corrosion pits that after 10 days of immersion

in 3.5 wt% NaCl solution decreases from 3.3~22.2  $\mu\text{m}$  to 153~260 nm, and the adhesion of the coating is increased from 20.5 N to 61.6 N.

The research results can provide theoretical and technical support for the preparation of new composite coatings with high efficiency, low cost, high adhesion and strong corrosion resistance.

**Keywords:** jet electrodeposition; composite coating; preferred orientation; adhesion force; corrosion resistance

## 1. Introduction

With the continuous development of modern industry, the wear resistance and corrosion resistance of more and more key components of high-end equipment are required, so the high-performance composite coatings are usually required to be prepared on the key component surface. Nanoparticles reinforced metal matrix composite coatings have great application potential in mechanical parts surface strengthening due to their excellent hardness, abrasion resistance, corrosion resistance and high temperature oxidation resistance. It is widely concerned by researchers. Ruiqian Li et al. [1] produced the Ni-SiO<sub>2</sub> composite coating by electrodeposition and found that the addition of SiO<sub>2</sub> nanoparticles improved the wear resistance of the coating. Baosong Li et al. [2] prepared the Ni-W/TiN coating by pulse electrodeposition and observed that the doped TiN nanoparticles could promote nucleation and caused obvious changes in microstructure, thus improving the hardness and corrosion resistance of the coating. Yongqi Tao et al. [3] fabricated the Ni-B-Sc coating by conventional electrodeposition and detected that the grain boundary and phase boundary area increased due to the addition of Ni<sub>2</sub>Sc nanoparticles. However, nanoparticles usually exist in the form of agglomeration in the plating solution because of their high surface energy. It is difficult to break the agglomeration of nanoparticles by conventional electrodeposition, so the prepared composite coatings have defects such as rough surface and poor adhesion [4, 5].

Jet electrodeposition (JED) is a kind of unconventional electrodeposition. The

high-speed jet liquid improves the transmission speed of ions in the deposition process compared with conventional electrodeposition (CED). The ions are evenly distributed by high-speed flushing, which reduces the concentration polarization and improves the upper limit value of current density in the process of electrodeposition [6, 7]. Meanwhile, the agglomeration of the nanoparticles is broken up during the high-speed flushing, and the nanoparticles are distributed uniformly in the coating. Therefore, using jet electrodeposition to prepare nanoparticles reinforced composite coating, the advantages of high deposition efficiency, uniform distribution of nanoparticles and good surface quality of coating can be obtained [8-10].

During electrodeposition, the preferred orientation (texture) often occurs, which means significant amounts of grains exhibit the same common orientation characteristics in the deposition layer. It is called highly preferred orientation if almost all of grains are assembled in one certain orientation. Through controlling the preferred orientation of the grains in the deposition layer, the properties of deposition layer can be improved, and even deposition layer has some special functions. Haibo Gao et al. [11] made Co-Ni films with preferred (220) orientation by controlling the variation of Co and Ni ratio, which showed superior photocatalytic performance. Alexandre Ponrouch et al. [12] prepared preferentially oriented (100) platinum nanowires and thin films by changing the deposition potential, which exhibited efficient electro-catalytic properties. Jiwon Kim et al. [13] manufactured  $\text{Bi}_2\text{Te}_{3-x}\text{Se}_x$  thin films by pulse electrodeposition. Through changing the duty cycle of current, thin films had the preferred orientation of (110), and displayed superior thermoelectric performance.

In recent years, the research on the preferred orientation of nickel coatings by conventional electrodeposition has attracted much attention. J.A. Calderón et al. [14] carried out the experiment about preparing Ni-SiC composite coatings. It was found that with the increase of SiC content, the grain orientation of the coatings gradually evolved from (200) to (111), and corrosion resistance of the coating was improved with the grain orientation of (111). Yuantao Zhao et al. [15] performed the research on  $\text{Ni-xAl-yTi}$  composite coatings. They claimed that as the content of Al and Ti particles increased, the orientation of (200) decreased while (111) increased, and the preferred

(111) orientation coating showed better corrosion resistance than preferred (200) orientation. Morteza Alizadeh et al. [16] undertook the analysis about Ni-Cu/Al<sub>2</sub>O<sub>3</sub> composite coatings. They reported the structure of coatings gradually evolved to preferred (111) orientation when the content of Al<sub>2</sub>O<sub>3</sub> increased, and the hardness, wear resistance and corrosion resistance of coatings were improved. Jianhua Deng et al. [17] demonstrated that with the addition of 1,4-bis(2-hydroxyethoxy)-2-butyne (BEO), the grains preferred orientation of Ni/diamond composite coatings changed to (200), and the wear resistance of the coating was also enhanced.

The above studies on the preferred orientation of nickel-based composite coatings mainly focused on (111) and (200), and research about the highly preferred (220) orientation of Ni-SiC composite coatings has not been reported. This paper reveals the principle of efficient preparation nanoparticles reinforced nickel-based composite coating with highly preferred (220) orientation, and then explores the effect of deposition parameters on the structure of nanoparticles reinforced nickel-based composite coatings, finally investigates the microstructure, corrosion resistance and the adhesion force of the Ni-SiC composite coating with highly preferred (220) orientation. Some innovative theoretical and technological achievements have been made.

## **2. Experimental**

### **2.1 Principle of efficient preparation of nanoparticles reinforced nickel-based composite coating by jet electrodeposition (JED).**

The experimental device for jet electrodeposition is shown in **Figure 1a**. The pure titanium rod connects to the positive pole of DC power supply and the substrate connects to the negative pole of DC power supply. The pure titanium rod acts as the current transfer electrode, which transfers the current to nickel balls, making nickel balls become anode. Nickel atoms are oxidized into a large amount of Ni<sup>2+</sup>, which are used to supplement the Ni<sup>2+</sup> constantly consumed in the plating solution. The titanium rod is not consumed in experiments because the chemical properties of titanium rod is more stable than those of nickel balls. Due to the titanium rod is closer to the upper

surface of the substrate, the unconsumed titanium rod can play a role in providing a stable electric field environment, and the deposited coatings have better uniformity. The plating solution in the liquid storage tank is heated by a water bath to maintain a constant temperature. During the preparation process, firstly the composite plating solution is transported to the anode cylinder through the inlet pipe by a diaphragm pump. Then the plating solution in the anode cylinder is impacted on the substrate through the nozzle at a high speed for deposition. Finally, the plating solution flows back to the liquid storage tank through the outlet pipe.

The schematic diagram of preparation principle of nanoparticles reinforced nickel-based composite coatings by JED is illustrated in **Figure 1b**. Most of the nanoparticles exist in the plating solution as agglomeration, which mainly are surrounded by water molecules and ion clusters. Since mainly chemical reaction is the redox of  $\text{Ni}^{2+}$  during electrodeposition, so the model is simplified into nanoparticles agglomeration surrounded by a large number of  $\text{Ni}^{2+}$ . When the nanoparticles agglomeration is impacted the substrate at a high speed, the nanoparticles agglomeration are divided into many individual nanoparticles due to reacting force. According to the composite co-deposition theory [18, 19], some nanoparticles are adsorbed on the substrate, among which the strongly adsorbed nanoparticles are deposited and the weakly adsorbed nanoparticles are washed away. Meanwhile, under the condition of high deposition current density, a large amount of  $\text{Ni}^{2+}$  near the cathode are reduced to Ni atoms, and then Ni atoms are deposited on the substrate. The stacking of Ni atoms and nanoparticles covering substrate defects rapidly, and eventually the nanoparticles reinforced nickel-based composite coating with smooth surface is formed.

As shown in **Figure 1c**, the thickness of the coating increase gradually when the nozzle moves back and forth along the path at a certain scanning speed. The required thickness and shape of the composite coating has been obtained by controlling the nozzle path and deposition time.

(Location of [Figure 1](#))

## 2.2 Experimental parameters of preparation of nanoparticles reinforced nickel-

### **based composite coating by JED.**

Because of the high hardness and stability of SiC nanoparticles, Ni-SiC composite coatings have been widely used. This paper focused on the properties of Ni-SiC composite coatings. The composition of plating solution and experimental parameters are shown in **Table 1**. The purity of SiC nanoparticles is 99.99% and the average particle size is about 50 nm, and the SEM and EDS figures are shown in **figure S1**.

(Location of [Table 1](#))

### **2.3 Testing details.**

The grain orientation and grain size of nanoparticles reinforced nickel-based composite coatings are characterized by X-ray diffraction (XRD, X'Pert Pro MPD) with Cu-K $\alpha$  ( $\lambda=0.154184$  nm) radiation, and the test parameters are voltage of 45 kV, current of 40 mA, scanning range of 20°~90°, step width of 0.01° and scanning speed of 10°/min. The adhesion force of nanoparticles reinforced nickel-based composite coatings are quantitatively tested by scratch tester (WS-2004). The test is carried out in the form of dynamic loading. The scratch length is 4 mm, and the diamond tool is uniformly loaded from 0~70 N at a loading rate of 70/4 N·mm<sup>-1</sup>. The morphology and element content of nanoparticles reinforced nickel-based composite coatings are measured by scanning electron microscope with energy dispersive spectroscopy (SEM, EDS, ZEISS MERLIN Compact). The surface roughness of nanoparticles reinforced nickel-based composite coatings are measured by TR300 Roughness Measuring Instrument and the sampling length is 4 mm. The electrochemical properties of Ni-SiC composite coatings are performed by electrochemical workstation (CHI 760e) in 3.5 wt% NaCl solution at the room temperature. The test samples are Ni-SiC composite coatings with an area of 1 cm<sup>2</sup>. A saturated calomel electrode, platinum plate and

samples are used as the reference electrode, counter electrode and working electrode, respectively. The polarization curves are recorded through the potential range of -150 mV to +350 mV (compared to the  $E_{ocp}$ ) at a scan rate of 0.002 V/s and a scan frequency of 2 Hz. The electrochemical impedance spectroscopy (EIS) investigations are generated in the sinusoidal signal amplitude of 10 mV and at a frequency range of 10<sup>5</sup> ~10<sup>-2</sup> Hz.

### 3. Results and discussion

#### 3.1 The grain orientation evolution of Ni-SiC composite coating during JED.

In the process of JED, the deposition layer is rapidly formed on the surface of the substrate with the condition of high current density by means of high-speed jetting. Therefore, the parameters of injection velocity and current density are important factors influencing the structure of the coatings. In this paper, a large number of Ni-SiC composite coatings are produced at different current densities and different injection velocities. The thicknesses of these coatings are all controlled to be about 50  $\mu\text{m}$  by changing the deposition time. According to **Figure 2a-2e**, the XRD patterns of Ni-SiC composite coatings prepared by jet electrodeposition under different deposition parameters all present clear diffraction peaks of Ni(111), Ni(200), and Ni(220). It can be seen that with the increase of the deposition current density from 180 A/dm<sup>2</sup> to 220 A/dm<sup>2</sup>, the coating orientation shows a trend of gradual evolution to Ni(220).

The preferred orientation coefficient (texture coefficients) of different crystal planes in coating of these coatings are calculated by **formula (1)** [20], where  $TC(hkl)$  is the texture coefficient of (hkl) orientation,  $I_{(hkl)}$  is measured intensity of (hkl) reflection,  $I_{0(hkl)}$  is powder diffraction intensity of nickel (PDF#70-0989), and  $n$  is the number of reflections used in the calculations. In this case, (111), (200) and (220) peaks were used for texture coefficient calculation ( $n = 3$ ).

$$TC(hkl) = \frac{I_{(hkl)} / I_{0(hkl)}}{\sum_{i=1}^n I_{(hkl)} / I_{0(hkl)}} \quad (1)$$

**Figure 2f-2h** demonstrates the variation of TC(111), TC(200) and TC(220) of Ni-SiC composite coatings prepared by jet electrodeposition under different deposition parameters. With the current density increases, it shows a general trend that TC(111) and TC(200) gradually decreases and TC(220) gradually increases. At low current density conditions (180 A/dm<sup>2</sup>-200 A/dm<sup>2</sup>), the TC(111), TC(200) and TC(220) are greatly affected by variation of injection velocity. As the injection velocity increases, TC(111) and TC(200) decreases first and then increases, and TC(220) increases first and then decreases. However, at a higher deposition current density (> 200 A/dm<sup>2</sup>), the grain orientation of the coatings are little affected by variation of injection velocity, and the TC(111), TC(200) and TC(220) remains stable value. Under the deposition conditions of 220 A/dm<sup>2</sup> current density and 1.76 m/s injection velocity, TC(220) reaches a maximum of 97.7%.

The grain sizes of these coatings are calculated by **formula (2)** [21], where D(hkl) represents grain size of (hkl) orientation, B represents half-height width of diffraction peak (degree),  $\gamma$  represents the wavelength of Cu-K $\alpha$  ( $\lambda=0.154184$  nm),  $\theta$  represents Bragg angle (rad) and K represents the constant (K = 0.89).

$$D(hkl) = \frac{K\gamma}{B \cos \theta} \quad (2)$$

As shown in **Figure 2i**, it can be seen that the variation range of D(111) and D(200) is small with the current density increased. D(111) is between 19~20 nm and D(200) is between 17~18 nm. Unlike D(111) and D(200), D(220) increases with the increase of deposition current density. When the deposition current density is 180 A/dm<sup>2</sup>, the

average value of  $D(220)$  is 47.5 nm. As the deposition current density increases to 220 A/dm<sup>2</sup>, the average value of  $D(220)$  increases to 61.6 nm.

(Location of [Figure 2](#))

As shown in **Figure 3**, in the process of preparing nanoparticles reinforced nickel-based composite coatings by jet electrodeposition, Ni atoms are stacked with face centered cubic structure cells. Usually, those cells are deposited on the substrate surface with (111) crystalline plane, (200) crystalline plane, and (220) crystalline plane. The selection of grains orientation mainly depends on cathodic overpotential, which is mainly determined by the deposition current density according to the Tafel formula. Under the condition of low deposition current density, the grain orientation of coating tended to (111). With the deposition current density increasing, the grain orientation of coating gradually evolves to (220). When the deposition current density exceeds a certain value, most of the cells are adsorbed on the substrate with the (220) crystalline plane.

(Location of [Figure 3](#))

Compared with the influence of deposition current density, the effect of injection velocity on the grain orientation of the coating is smaller. Because the bonding strength of (111) and (200) oriented grains to the substrate is less than that of (220) oriented grains (**Chapter 3.4**), the increase of injection velocity makes the nickel structure cells tend to adsorb on the substrate with (220) crystalline plane. However, the  $TC(220)$  of the coating does not increase monotonically with the increase of the injection velocity. There is a critical value for the injection speed. When the injection speed exceeds this value, the nickel structure cells tend to adsorb on the substrate with random crystalline planes instead. The critical value has a negative correlation with the deposition current density, that is, the smaller the deposition current density, the larger the critical value (**Figure 2f-2h**).

Under the condition of high deposition current density, the nickel structure cells nucleate rapidly, and the number of crystal nucleus with (220) orientation is larger than that of with (111) orientation and (200) orientation. Therefore, the crystal nucleus with

(220) orientation dominate the growth process. Eventually, the average size of (220) orientation grains is obviously larger than that of (111) orientation grains and (200) orientation grains (**Figure 2i**).

In order to explore the effect of nanoparticles in the preparation of nickel-based composite coatings with highly preferred (220) orientation, the pure Ni coating (**Figure S2-S3**), Ni-TiO<sub>2</sub> composite coating (**Figure S4-S5**) and Ni-Al<sub>2</sub>O<sub>3</sub> composite coating (**Figure S6-S7**) are also prepared by JED. The deposition current density is 220 A/dm<sup>2</sup> and the injection velocity is 1.76 m/s. The results show that all of the three coatings are all highly preferred (220) orientation structure, which shows that nanoparticles have little effect on the orientation of the grains in the coating. This paper will carry out subsequent analysis based on Ni-SiC composite coating.

### **3.2 The morphology of Ni-SiC composite coating with highly preferred (220) orientation.**

**Figure 4** shows the morphology of a Ni-SiC composite coating with highly preferred (220) orientation prepared by JED (TC(220) = 97.7%). As shown in **Figure 4a**, the surface (Area C) and section (Area f) of the coating are observed. It is obvious that the surface of Ni SiC composite coating with highly preferred (220) orientation is compact and flat (**Figure 4c**, **Figure 4f**), and there is no dome-like or hill valley like structure [16, 22-24]. The surface roughness of the coating is Ra0.119 μm (**Figure 4b**). SiC nanoparticles have no agglomeration inside the coating (**Figure 4d**, **Figure 4g**) and are evenly distributed in different areas of the coating (**Figure 4e**, **Figure 4h**). **Figure 5** shows the morphology of a Ni-SiC composite coating prepared by CED (**Figure S8**, **Table S1**). The XRD pattern of the coating is shown in **Figure S9** (TC(220) = 40.7%).

(Location of [Figure 4](#))

As shown in **Figure 5a**, the surface (Area C) and section (Area f) of the coating are also observed. Unlike **Figure 4c** and **Figure 4f**, the surface of Ni SiC composite coating prepared by CED is coarse and fluctuant (**Figure 5c**, **Figure 5f**). The surface roughness of the coating is Ra1.210 μm (**Figure 5b**). SiC nanoparticles have obvious agglomeration inside the coating (**Figure 5d**, **Figure 5g**) and the content varies greatly

at different areas (**Figure 5e, Figure 5h**).

(Location of **Figure 5**)

The high speed jetting process in JED can make SiC nanoparticles evenly distributed in the coating. At the same time, the highly preferred (220) orientation structure makes the coating has better surface quality, which is beneficial to significantly improve the corrosion resistance of Ni-SiC composite coating.

### **3.3 The corrosion resistance of Ni-SiC composite coating with highly preferred (220) orientation.**

In this paper, the corrosion resistance of Ni-SiC composite coating is tested, including electrochemical test and full immersion corrosion test. **Figure 6** reports the electrochemical test results of Ni-SiC composite coatings in 3.5 wt% NaCl solution. The dynamic polarization curves are shown in **Figure 6a**. As can be seen from this figure, the corrosion potential ( $E_{\text{corr}}$ ) and corrosion current density ( $i_{\text{corr}}$ ) of the Ni-SiC composite coating prepared by CED are -0.747 V and  $54.52 \mu\text{A}\cdot\text{cm}^2$ , respectively.

The Ni-SiC composite coatings are prepared by JED at an injection velocity of 1.76 m/s and a current density of 180~220 A/dm<sup>2</sup>. With the increase of the deposition current density, the TC(220) of the coating increases from 41.4% to 97.7%, the corrosion potential of the coating increases from -0.575 V to -0.477 V, and the corrosion current density decreases from  $9.52 \mu\text{A}\cdot\text{cm}^2$  to  $2.76 \mu\text{A}\cdot\text{cm}^2$ . This indicates that the corrosion resistance of the coating is enhanced. Electrochemical impedance spectroscopy (EIS) tests are also conducted and results are shown in **Figure 6b-6d**. **Figure 6b** shows the typical Nyquist plots of the Ni-SiC composite coatings. A depressive semicircle can be found on each curve, indicating that the electrochemical mechanism is controlled by the charge-transfer process [25]. It can be seen that the diameter of capacitive loop increases with the increases of TC(220) of Ni-SiC composite coating. The larger the diameter of the capacitive loop, the stronger the corrosion resistance of the coating. **Figure 6c** shows the Bode plots of  $\log(f)$  vs.  $\log(|Z|)$  of the Ni-SiC composite coatings. In the high-frequency region ( $10^4\sim 10^5$  Hz), the values of  $\log|Z|$  are similar, indicating that these are impedance values of 3.5 wt% NaCl

solution. In the low-frequency region ( $10^{-2}$ ~ $10^{-1}$  Hz), the impedance values of different Ni-SiC composite coatings are significantly different. For instance, at the fixed 0.01 Hz, the  $\log(|Z|)$  of Ni-SiC composite coating prepared by CED is 3.41, and the  $\log(|Z|)$  of Ni-SiC composite coating prepared by JED increase from 3.99 to 4.72 as the TC(220) increase from 41.4% to 97.7%. The higher the impedance value, the stronger the corrosion resistance of the coating. **Figure 6d** shows the Bode plots of  $\log(f)$  vs. Angle of the Ni-SiC composite coatings. For the Ni-SiC composite coatings, the higher phase angle at middle high frequency for the chemical conversion treated specimen corresponds to a capacitive behavior, that is to say the conversion coating has good dielectric property to avoid the ionic flow of electrolyte [25]. When the frequency is in the range of  $10^0$ ~ $10^5$  Hz, the phase angle of Ni-SiC composite coating prepared by CED is minimum, and the phase angle of Ni-SiC composite coating prepared by JED increases with the increase of TC(220).

The EIS data is fitted by calculated by electrical equivalent circuit (EEC). The corrosion process of Ni-SiC composite coating can be replaced by EEC of  $R(Q(R(QR)))$  when the oxide layer on the substrate surface is taken into account [26]. As observed in **Figure 6e**, The EEC is consisted of the  $R_s$  modeling the solution resistance, in sequence with a constant phase element  $CPE_1$  in parallel with another resistance  $R_c$  modeling the coating resistance, and then another constant phase element  $CPE_2$  and the third charge transfer resistance of  $R_{ct}$  associating with the corrosion process. The impedance of CPE is defined as  $Q_{CPE} = [Y_0(j\omega)^n]^{-1}$ , where  $\omega$  is the angular frequency (rad s<sup>-1</sup>),  $Y_0$  is the CPE admittance,  $j$  is the imaginary number ( $\sqrt{-1}$ ), and  $n$  with a value of 0-1 represents the relaxation dispersion. When the value of  $n$  is 1, the CPE is a pure capacitor with a capacitance of  $Y_0$ . It is believed that the smaller the  $n$  value is and the more defects will be on the surface as well as the pitting corrosion is more likely to occur [25, 27].

(Location of [Figure 6](#))

The fitting results is shown in **Table 2**. It can be seen that the  $R_c$  and  $R_{ct}$  of Ni-SiC composite coating prepared by CED is  $1.476 \times 10^2 \Omega/\text{cm}^2$  and  $1.476 \times 10^2 \Omega/\text{cm}^2$ , respectively. Compared with that of other coatings, these parameters is the minimum

value, which means this coating has the worst corrosion resistance. The Ni-SiC composite coatings prepared by JED increase with TC (220) from 41.4% to 97.7%, the  $R_c$  of the coatings increase from  $4.423 \times 10^3 \Omega/\text{cm}^2$  to  $7.025 \times 10^3 \Omega/\text{cm}^2$ , and the  $R_{ct}$  increase from  $1.267 \times 10^4 \Omega/\text{cm}^2$  to  $5.989 \times 10^4 \Omega/\text{cm}^2$ . It is proved that the corrosion resistance of Ni-SiC composite coating increases with the increase of TC(220).

(Location of [Table 2](#))

The full immersion corrosion tests of Ni-SiC composite coatings are carried out. The corrosion solution was 3.5 wt% NaCl solution, and the corrosion time was 1 day, 3 days, 5 days and 10 days, respectively. The surface of the coatings after immersion corrosion are observed, and the surface morphologies of different corrosion times are shown in **Figure S10-S12** and **Figure 7**.

**Figure 7a** is the surface morphology of Ni-SiC composite coating prepared by CED after 10 days of immersion corrosion. It can be found that many corrosion products adhere to the surface of the coating, and the size of corrosion pits is about 3.3~22.2  $\mu\text{m}$ . **Figure 7b-7f** are the surface morphologies of Ni-SiC composite coatings prepared by JED after 10 days of immersion corrosion. Compared with **Figure 7a**, there are no obvious corrosion products on the surface of the coating, and the corrosion pits are all less than 1  $\mu\text{m}$ . When the TC(220) of the coating is 41.4%, the size of corrosion pits is about 278~944 nm (**Figure 7b**). When the TC(220) of the coating is 63.9%, the size of corrosion pits is about 254~927 nm (**Figure 7c**). When the TC(220) of the coating is 75.6%, the size of corrosion pits is about 215~905 nm (**Figure 7d**). When the TC(220) of the coating is 93.4%, the size of corrosion pits is about 183~352 nm (**Figure 7e**). When the TC(220) of the coating is 97.7%, the size of corrosion pits is about 153~260 nm (**Figure 7f**). This shows that as the TC(220) increases, the corrosion resistance of the Ni-SiC composite coating is improved.

(Location of [Figure 7](#))

Surface quality is the most important factor affecting the corrosion resistance of the coating. The corrosion resistance of the coating decreases with the increase of the number of surface defects. There are many defects on the surface of Ni-SiC composite

coating prepared by CED, such as the agglomeration of SiC nanoparticles, pinholes and pockmarks. Corrosion occurs preferentially at these defect locations, and over a period of time, it is easy to cause SiC agglomerated particles to fall off, resulting in the formation of holes in the coating, which eventually form larger corrosion pits with the increase of corrosion time. The surface of Ni-SiC composite coatings prepared by JED are compact and flat without obvious defects and SiC particles agglomeration, so the corrosion resistance of coatings are significantly improved. The corrosion pits occur uniformly at the grain boundaries on the coating surface, and the corrosion pits gradually expanded with the increase of corrosion time. Under the same corrosion area, the smaller the grain boundaries proportion and the less the number of corrosion pits. With the increase of deposition current density, the grain size of Ni-SiC composite coating also increases (**Figure 2f**), and the proportion of grain boundaries decreases. At the same time, with the increase of TC(220), the coating with the same orientation gradually formed. The dislocations and other defects at grain boundaries are also reduced and the diffusion rate of corrosion pits is reduced. Therefore, the Ni-SiC coating with highly preferred (220) orientation shows excellent corrosion resistance.

#### **3.4 The adhesion force of Ni-SiC composite coating with highly preferred (220) orientation.**

The adhesion force is an important index to judge the performance of coating. The higher the adhesion force of the coating, the less easy it will fall off, and the longer the service life will be. In this paper, the adhesion of Ni-SiC composite coatings and substrate is quantitatively measured by scratch method. **Figure 8a** shows the scratch morphology of Ni-SiC composite coating prepared by CED, and **Figure 8b-8f** show the scratch morphologies of Ni-SiC composite coatings prepared by JED under the deposition current density of 180~220 A/dm<sup>2</sup>. As shown in the elliptical frame selection area, the load corresponding to the first crack position of the coating is defined as the value of adhesion force, which is 20.5 N、24.9 N、46.8 N、55.2 N、60.5 N and 61.6N, respectively. It can be seen that the adhesion force of Ni-SiC composite coating has a positive correlation with the value of TC(220). The higher TC (220), the higher the

adhesion force of the coating.

From the microscopic point of view, the adhesion force between coating and substrate depends on the contact strength of interface. A fundamental quantity determining the strength of interface is the ideal work of adhesion ( $W_{ad}$ ), which is the work spent on separation the interface into two free surface. The work of adhesion between coating and substrate can be calculated by the **Formula (3)** [28, 29].

$$W_{ad} = (E_{slab1} + E_{slab2} - E_{int}) / A \quad (3)$$

Where  $E_{slab1}$  and  $E_{slab2}$  are the total energies of slab1 and slab2,  $E_{int}$  is the total energy of the interface system including slab1 and slab2, and  $A$  is the interface area.

The DFT simulation models are shown in **figure 8g**. The substrate is medium carbon steel, the iron content is 98.39 wt% (**Figure S13**), and the main orientation of the substrate is Fe (110) (**Figure. S14**). The  $W_{ad}$  of Ni(111)/Fe(110), Ni(200)/Fe(110), and Ni(220)/Fe(110) interfaces are 3.32J/m<sup>2</sup>, 3.96J/m<sup>2</sup> and 4.21J/m<sup>2</sup>, respectively. The  $W_{ad}$  of Ni(220)/Fe(110) interface is more than that of Ni(111)/Fe(110) interface and Ni(200)/Fe(110) interface, indicating that the adhesion force between Ni-SiC composite coating with highly preferred (220) orientation and substrate is higher.

(Location of [Figure 8](#))

## 4. Conclusions

(1) The grain orientation of Ni-SiC composite coatings prepared by JED gradually evolves to (220) with the increase of current density. The Ni-SiC composite coating with highly preferred (220) orientation is prepared at current density of 220 A/dm<sup>2</sup>, while the orientation coefficient reached to 97.7%.

(2) In the process of JED, the high-speed jetting fluid makes the nanoparticles break the agglomeration state and disperse evenly into the coating. Under the condition

of high current density, the deposition layer quickly to fill the defects of the substrate and form a compact and flat composite coating on the surface. The surface roughness of Ni-SiC composite coating with highly preferred (220) orientation is 90.2% lower than that of Ni-SiC composite coating prepared by CED.

(3) The corrosion resistance of Ni-SiC composite coatings increases with the increase of TC(220). Compared with the corrosion resistance of coating prepared by CED, the corrosion resistance of coating with highly preferred (220) orientation prepared by JED has a 36.1% increase in corrosion potential and a 94.9% decrease in corrosion current density.

(4) DFT simulation is used to calculate the ideal work of adhesion of Ni(111)/Fe(110) interface, Ni(200)/Fe(110) interface, and Ni(220)/Fe(110) interface of the Ni-SiC composite coating, among which Ni(220)/Fe(110) interface has higher  $W_{ad}$  than Ni(111)/Fe(110) interface and Ni(200)/Fe(110) interface. The Ni-SiC composite coating with a highly preferred (220) orientation obtains the maximum adhesion force, and the adhesion force is 200% higher than that of the Ni-SiC composite coating prepared by CED.

## **Declarations**

## **Abbreviations**

**JED**: Jet electrodeposition; **CED**: Conventional electrodeposition; **TC**: Texture coefficient; **SEM/EDS**: Scanning electron microscope/Elemental dispersive X-ray spectrometry; **XRD**: X-ray diffraction; **EIS**: Electrochemical impedance spectroscopy; **EEC**: electrical equivalent circuit.

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## **Authors' contributions**

Renjie Ji and Hui Jin contributed equally. The manuscript was written by Renjie Ji, Yonghong Liu and Hui Jin. Experiments were designed by Tiancong Dong, Fan Zhang, Lilong Zhao and Chi Ma. Xinlei Wu, Qiang Sun, Hang Dong, Dege Li, Peng Liu and Baoping Cai performed the experiments. All authors have given approval to the final version of the manuscript.

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## **Availability of data and materials**

All data supporting the conclusions of this article can be obtained from the corresponding authors upon reasonable request.

## **Declaration of interest**

The authors declare no competing financial interest.

## **Supporting information:**

The Supporting Information is available.

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## Figure Captions

**Figure 1.** Schematic diagram of (a) experimental device for JED, (b) preparation principle of nanoparticles reinforced nickel-based composite coatings, (c) production process of nanoparticles reinforced nickel-based composite coatings.

**Figure 2. (a-e)** XRD patterns of Ni-SiC composite coatings prepared by jet electrodeposition under different deposition parameters: (a) injection velocity of 1.40~2.65 m/s at 180 A/dm<sup>2</sup> current density, (b) injection velocity of 1.40~2.65 m/s at 190 A/dm<sup>2</sup> current density, (c) injection velocity of 1.40~2.65 m/s at 200 A/dm<sup>2</sup> current density, (d) injection velocity of 1.40~2.65 m/s at 210 A/dm<sup>2</sup> current density, (e) injection velocity of 1.40~2.65 m/s at 220 A/dm<sup>2</sup> current density. (f-h) The texture coefficients of Ni-SiC composite coatings prepared by jet electrodeposition under different deposition parameters: (f) TC(111), (g) TC(200), (h) TC(220). (i) The grain size statistic of Ni-SiC composite coatings prepared by jet electrodeposition under different deposition parameters.

**Figure 3.** Schematic diagram of the evolution of grain orientation in the process of preparing nanoparticles reinforced nickel-based composite coatings by JED.

**Figure 4.** The morphology of the Ni-SiC composite coating with highly preferred (220) orientation prepared by JED: (a) schematic diagram of detection area, (b) roughness profile of the coating surface, (c) microstructure of the coating surface, (d) distribution of SiC nanoparticles on the coating surface, (e) EDS map of the coating surface, (f) microstructure of the coating section, (g) distribution of SiC nanoparticles on the coating section, (h) EDS map of the coating section.

**Figure 5.** The morphology of the Ni-SiC composite prepared by CED: **(a)** schematic diagram of detection area, **(b)** roughness profile of the coating surface, **(c)** microstructure of the coating surface, **(d)** distribution of SiC nanoparticles on the coating surface, **(e)** EDS map of the coating surface, **(f)** microstructure of the coating section, **(g)** distribution of SiC nanoparticles on the coating section, **(h)** EDS map of the coating section.

**Figure 6.** Electrochemical test results of Ni-SiC composite coatings: **(a)** dynamic polarization curves, **(b)** Nyquist plots, **(c)** Bode plots of  $\log(f)$  vs.  $\log(|Z|)$ , **(d)** Bode plots of  $\log(f)$  vs. Angle, **(e)** schematic diagram of the EEC.

**Figure 7** The surface morphologies of different Ni-SiC composite coatings after 10 days of immersion corrosion in 3.5 wt% NaCl solution: **(a)** CED (8 A/dm<sup>2</sup>), **(b)** JED (180 A/dm<sup>2</sup>), **(c)** JED (190 A/dm<sup>2</sup>), **(d)** JED (200 A/dm<sup>2</sup>), **(e)** JED (210 A/dm<sup>2</sup>), **(f)** JED (220 A/dm<sup>2</sup>).

**Figure 8. (a-f)** Scratch morphology of different Ni-SiC composite coatings: **(a)** CED (8 A/dm<sup>2</sup>), **(b)** JED (180 A/dm<sup>2</sup>), **(c)** JED (190 A/dm<sup>2</sup>), **(d)** JED (200 A/dm<sup>2</sup>), **(e)** JED (210 A/dm<sup>2</sup>), **(f)** JED (220 A/dm<sup>2</sup>). **(g)** DFT optimized structures of Ni(111)/Fe(110), Ni(200)/Fe(110) and Ni(220)/Fe(110) interfaces.

## **Table Captions**

**Table 1** The composition of plating solution and experimental parameters.

**Table 2.** Parameters fitted from EEC.