



A study on the mechanical and thermal shock properties of MCrAlY coating prepared by arc ion plating

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ABSTRACT

In this study, MCrAlY coatings with different microstructures were prepared by arc ion plating to investigate the mechanical and thermal shock properties of the coatings. The results show that arc ion plating MCrAlY coatings overall possessed similar hardness (7.66 GPa–9.20 GPa) but lower elastic modulus (119.07 GPa–148.02 GPa), compared to those prepared by conventional thermal sprayings. The microstructure of the coating had a significant impact on the thermal shock property. The coating with a columnar structure exhibited a poor thermal shock property. After the thermal shock test, the coating failed in the form of penetrating cracks, which ultimately caused stress corrosion in the substrate. A dense and laminar structure was a desirable structure for ideal thermal shock property, which could be achieved by increasing the bias voltage applied to the substrate during the preparation process of the coating. However, in that case, the oxidation resistance of the coating was decreased due to the loss of Al in the coating.

1. Introduction

High-temperature protective coatings are widely applied in the blades or other hot components of gas turbines and aircraft engines. As a typical representative, MCrAlY coatings (where M = Co, Ni or Co/Ni) are largely used as a bonding layer of the thermal barrier coatings (TBCs). The bonding layer is an integral part of TBCs system. On the one hand, it can produce an effective protective oxide scale on the surface to prevent the oxidation of the substrate; On the other hand, it can also balance the thermal expansion coefficient difference between the top coating and the substrate, thereby improving the thermal shock resistance of TBCs. Up to now, the reported techniques used to prepare MCrAlY coatings mainly include air plasma spray [1,2], high-velocity oxy-fuel spray [3,4], vacuum plasma spray [5,6], and magnetron sputtering [7,8]. In recent years, arc ion plating (AIP) as a kind of PVD (physical vapour deposition) technique widely applied in the industry has been attracting more and more attention in terms of the preparation of the MCrAlY coatings [9–11]. The advantages of this technique are mainly reflected in the following aspects: firstly, vacuum preparation environment prevents the coating from being contaminated by

impurities during the preparation process, thus offering the coating a stronger adhesion as well as a higher oxidation resistance; secondly, overcoming the line-of-sight effect, which allows arc ion plating to deposit uniform coatings on the complex geometric component such as turbine blades; thirdly, fine microstructure of the resulting coating, which favours the formation of the protective Al₂O₃ scale on the coating surface at elevated temperatures [11]; finally, relatively high deposition efficiency, which makes arc ion plating more advantageous to apply in the industry compared to other PVD techniques such as magnetic sputtering.

The current reports concerning arc ion plating MCrAlY coatings are mainly focused on oxidation behaviour or hot corrosion behaviour. Researchers pay rare attention to their mechanical and thermal shock properties. The thermal shock property refers to the ability of the coating not to be destroyed under rapid temperature changes in cooling and heating. It is closely related to the mechanical properties (hardness and elastic modulus) of the coating itself. During the service of TBCs, there is a huge difference in thermal matching coefficient between ceramic coating and superalloy substrate. For that reason, MCrAlY coating as a bonding layer was commonly introduced between them. As

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a result, huge stress and strain tend to form at the interface of the bonding layer/ceramic layer or the bonding layer/superalloy substrate. Therefore, the mechanical properties of the bonding coating somehow play a crucial role in the thermal shock behaviour of TBCs system. Commonly speaking, the thermal sprayed MCrAlY coatings have superior thermal shock resistance. In that case, the coating spallation or even microstructural change is usually negligible after thermal shock [12,13]. However, in a limited number of reports on the arc ion plating MCrAlY coatings [14,15], the authors notice the remarkable change in the microstructure of the coatings after thermal shock, where a number of penetrating cracks occurred in the coating. This microstructural change implies that MCrAlY coatings prepared by arc ion plating may have a relatively poor thermal shock property. However, these reports did not pay sufficient attention to the observations above.

From this point of view, it is meaningful to figure out the failure mechanism of the arc ion plated MCrAlY coating after thermal shock. In this study, the mechanical properties and thermal shock properties of arc particle plating MCrAlY coatings were investigated in depth. The coatings with different microstructures were prepared by varying the bias voltage in order to evaluate the effect of coating microstructure on the thermal shock behaviour.

2. Experiment

2.1. Coating preparation

NiCoCrAlYTa alloy in size of $\Phi 100\text{mm} \times 40\text{ mm}$ was used as the targets, containing nominal chemical composition (wt%) of Co 20–25; Cr 19–21; Al 7–9; Y 0.5–1; Ta 1.5–3 and Ni Bal. Ni-based superalloy K423 A in size of $30 \times 10 \times 15\text{ mm}$ was used as the substrate, containing nominal chemical composition (wt%): Co 8.2–9.5; Cr 14–15.5; Al 3.9–4.4; Mo 6.8–8.3; Ti 3.4–3.8; W ≤ 0.20 ; Mn ≤ 0.20 ; C 0.12–0.18 and Ni Bal. A plating machine (AS-700, ProChina Limited, Beijing, China) was used to deposit the NiCoCrAlYTa coating. The schematic diagram of the plating machine is shown in Fig. 1. The substrates were successively treated by ground using No. 400# SiC abrasive paper, wet sand-blasted, ultrasonically cleaned, oven-dried and cleaned using acetone before being put in the plating machine. The vacuum chamber was evacuated to a base value of $2 \times 10^{-2}\text{ Pa}$; thereafter, it was maintained under 0.5 Pa by introducing argon. Prior to the coating deposition, an ion bombardment cleaning was carried out to remove surface contaminations and to

improve the surface activity of the substrate. The detailed deposition parameters are shown in Table 1.

2.2. Coating properties

The mechanical properties of the coatings were measured by means of nanoindentation (Hysitron, USA). The cross-section of the coatings was chosen as the measured position for the nanoindentation. The loading and unloading rates were 200 mN/min, and the pause time at the maximum load (100 mN) is 10 s. 10 different regions were measured to calculate the average values for each mechanical property.

Thermal shock test was carried out using a SX2-10-13 box-type resistance furnace with an accuracy of ± 1 . Since the research subject in this study is the bonding layer (MCrAlY coating) rather than intact TBCs, the temperature the MCrAlY coating can withstand is 100–150 °C lower than that of TBCs. Furthermore, the coating in this study is relatively thin (only 20–30 μm); thus, the requirements for the thermal shock property are relatively low as well. With these in mind, in this study, the coated samples were placed in the furnace remaining at the temperature at 950 °C for 5 min, and then quickly take them out and put them into water ($< 25\text{ °C}$). After the samples were thoroughly cooled, they were placed in the furnace again, thereafter repeating the above process 50 times.

2.3. Coating characterisation

The surface roughness (Ra) of the coatings was measured by a surface roughness tester (SJ-210 SERIES, Mitutoyo, Japan). Each coating was measured 10 times in different regions to calculate the average value. The microstructure of the coating was characterised by scanning electron microscopy (SEM) with energy-dispersive spectroscopy (Nova-Nona-430, FEI, Netherlands and JSM-7800F, JEOL, Japan). The phase composition of the coating was tested by X-ray diffraction (Bruker AXS D8 Focus, Germany) with $\text{CoK}\alpha$ radiation ($\lambda = 1.7889\text{ \AA}$). In order to protect the oxides scales from spalling during the preparation process of the samples, a layer of Al film was deposited on the coating by vacuum plating.

3. Result

3.1. Microstructure of the as-deposited coatings

Fig. 2 shows the surface SEM images of the coatings under different bias voltages. It can be observed that -50 V coating exhibits a cauliflower-like surface. As the bias voltage increases, the feature of the cauliflower gradually disappears, accompanied by a decrease of the surface roughness from 1.92 μm to 1.50 μm . As a result, -150 V coating shows a flat and compact surface, which significantly differs from the rough and loose surface of -50 V coating.

Fig. 3 shows the cross-sectional SEM images of the coatings under different bias voltages. It can be observed that -50 V coating possesses a columnar-like structure with high porosity. As the bias voltage increase, the coating became denser. As for -150 V coating, there is no obvious pore in the coating, and the coating shows a laminar structure similar to those prepared by thermal sprayings [16–18].

Table 1

The deposition parameters of the NiCoCrAlYTa coatings prepared by arc ion plating.

Bias voltage/V	–50, –100, –150
Working pressure/Pa	0.5
Arc current/A	110
Duty/%	70
Deposition time/h	4
Coating thickness/ μm	25–30

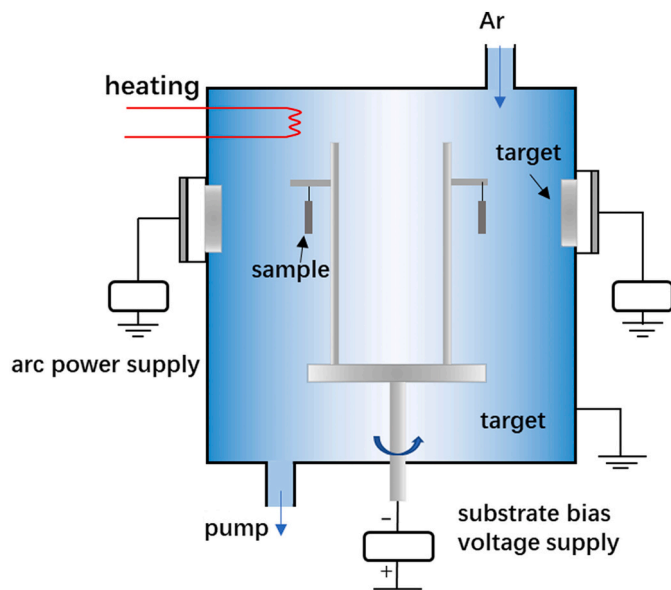


Fig. 1. Schematic diagram of arc ion plating in this study.

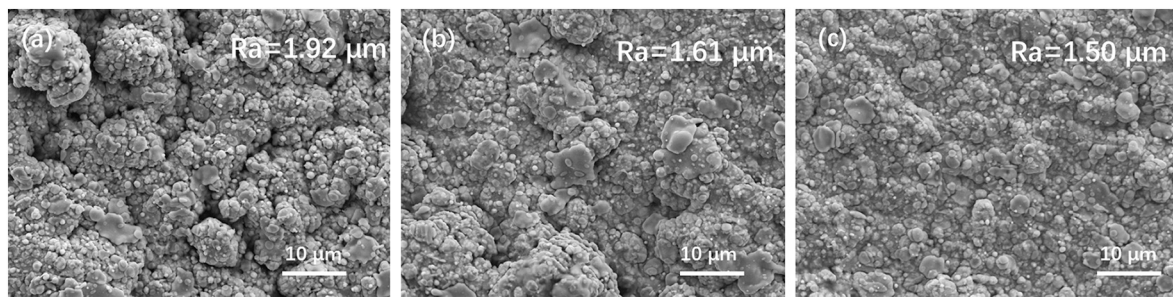


Fig. 2. Surface morphologies of the NiCoCrAlYTa coatings under different bias voltages: (a) - 50 V coating, (b) -100 V coating, (c) -150 V coating.

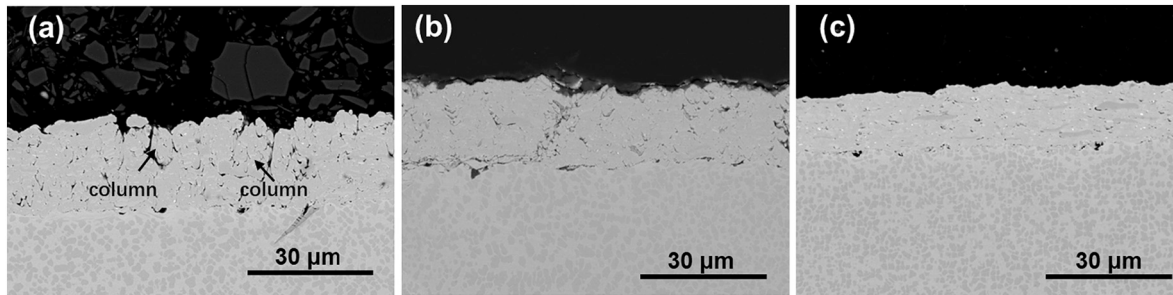


Fig. 3. Cross-sectional morphologies of the NiCoCrAlYTa coatings under different bias voltages: (a) - 50 V coating, (b) -100 V coating, (c) -150 V coating.

3.2. Mechanical properties of the as-deposited coatings

The mechanical properties of the as-deposited coatings are shown in Fig. 4. It can be seen that all the coatings display a smooth load-displacement curve (Fig. 4a). The maximum loading depth of -50 V, -100 V, -150 V coating is 890.63 nm, 805.77 nm and 786.07 nm, respectively. Fig. 4b shows the result of elastic modulus and hardness based on the indentation test. Both of them are roughly increased as the bias voltage increases (Fig. 4b). This result is in good agreement with the load-displacement curve of the indentation in Fig. 4a. Commonly speaking, the hardness and the elastic modulus of the coating strongly depend on its porosity [19]. The higher the porosity is, the lower the hardness or elastic modulus is. Since the coating porosity decreases with the increase in bias voltage, the coating under a higher bias voltage has a higher hardness and elastic modulus. It is worth noting that the hardness and the elastic modulus of the coating in this study are 7.66 GPa–9.20 GPa, 119.07 GPa–148.02 GPa, respectively. However, Han et al. [20] reported the mechanical properties of MCrAlY coatings prepared by plasma spraying and high-velocity air flame (HVOF), the result showed that the coating hardness ranged in 6 GPa–10 GPa, and the elastic modulus

ranged in 150 GPa–300 GPa. The above result indicates that arc ion plating MCrAlY coatings overall possess similar hardness but lower elastic modulus compared to those prepared by conventional thermal sprayings. The low elastic modulus can help to release the stress and strain during thermal shock, thereby preventing the appearance of crack [19,21]. From this perspective, arc ion plating has a significant advantage in preparing MCrAlY coatings. The lower elastic modulus of arc ion plating MCrAlY coating should be relevant to the finer microstructure. As shown in Fig. 2, the particle size of the arc ion plating NiCoCrAlYTa coating is mostly 1–2 μm. In contrast, that of the coating prepared by conventional thermal sprayings are mostly beyond 10 μm due to the limitation of the feed powder size. The large number of interparticle interfaces in the arc ion plating coating is beneficial to the localised deformation, thus lowering the elastic modulus of the coating.

3.3. XRD patterns of the coatings before and after the thermal shock test

Fig. 5 displays the phase composition of the coatings before and after the thermal shock. For the as-deposited coatings, there is no noticeable difference in the XRD pattern. All the coatings only consist of γ' -NiAl/

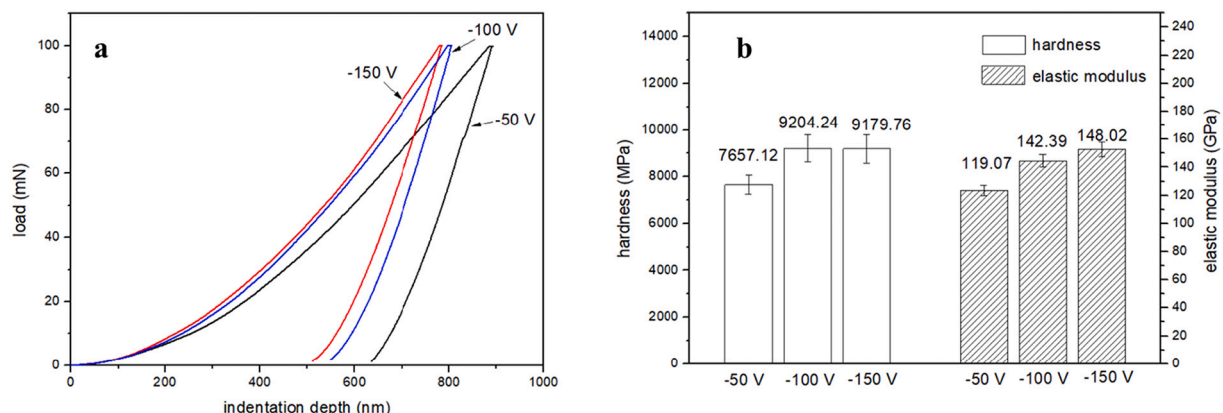


Fig. 4. Mechanical properties of the coatings: (a) typical load-displacement curves of nanoindentations; (b) hardness and elastic modulus of the coatings.

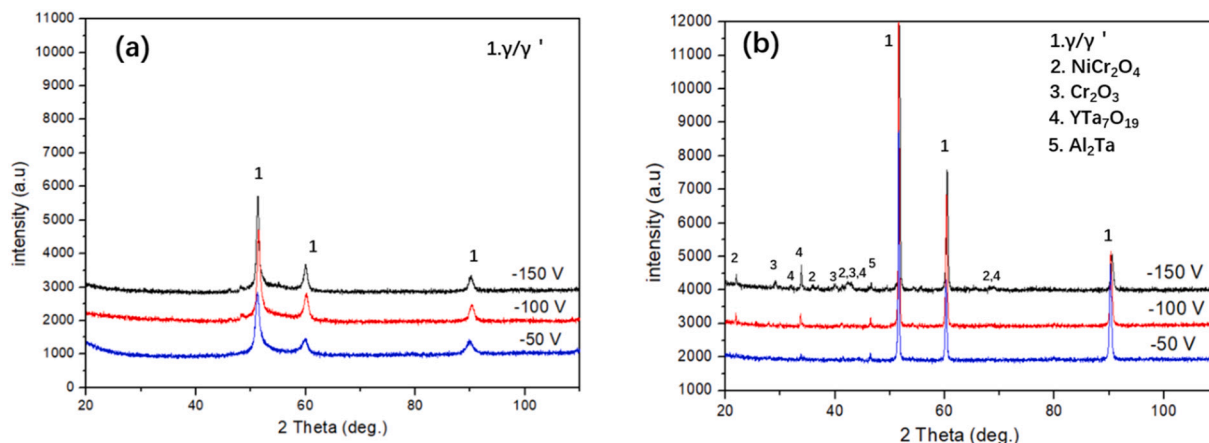


Fig. 5. XRD patterns of the NiCoCrAlYTa coatings under different bias voltages: (a) as-deposited coatings, (b) coatings after thermal shock.

γ -Ni. It indicates that the bias voltage has a negligible influence on the phase composition of the coating. After the thermal shock, the diffraction peaks of γ '-NiAl/ γ -Ni are much stronger and sharper than those of the as-deposited coatings, indicating that the growth of the grains in the coating during the thermal shock process. Here, Scherrer Eq. (1) is introduced to roughly calculate the average grain of the coatings, as shown below. Where D is the average grains size; $K = 0.89$; λ is the X-ray wavelength; β is the full width at half maximum (FWHM) of the diffraction peak, θ is the diffraction angle.

$$D = \frac{K\lambda}{\beta \cos \theta} \quad (1)$$

The average grain size of the as-deposited coating under -50 V, -100 V and -150 V is calculated to be 34.41 nm, 30.45 nm and 26.53 nm, respectively. After the thermal shock, they are increased to 83.34 nm, 79.95 nm and 69.29 nm, respectively. In addition to the grain growth, some reactive oxides (NiCr_2O_4 , Cr_2O_3 , $\text{YTa}_7\text{O}_{19}$) are detected on the surface of the coatings after thermal shock. Moreover, with the increase in bias voltage, the diffraction peaks of these oxides are

intensified to a certain extent.

3.4. Microstructure of the coatings after the thermal shock test

Fig. 6 shows the surface SEM images of the coatings after thermal shock. No notable spallation occurs on the surface of all the samples. However, a number of cracks exist on both -50 V coating and -100 V coatings (Fig. 6a, b). Furthermore, -50 V coating shows more cracks than -100 V coating. At higher magnification (Fig. 6a'–c'), it can be observed that the cracks mainly propagate along the notch of the coating surface morphology. Moreover, the cracks are much broader and deeper in the case of the coating under a lower bias voltage. In addition to the cracks, the oxide scales on the surface of the coatings also show considerable variation. In details, only Al_2O_3 was formed on -50 V coating. However, as bias voltage increases, more spinel oxides are formed on the coating. As a result, the surface of -150 V coating is fully covered by spinel oxides. This observation is well consistent with the XRD pattern of the coatings in Fig. 5, where the diffraction peaks of spinel oxides (mainly composed of NiCr_2O_4) are intensified with the

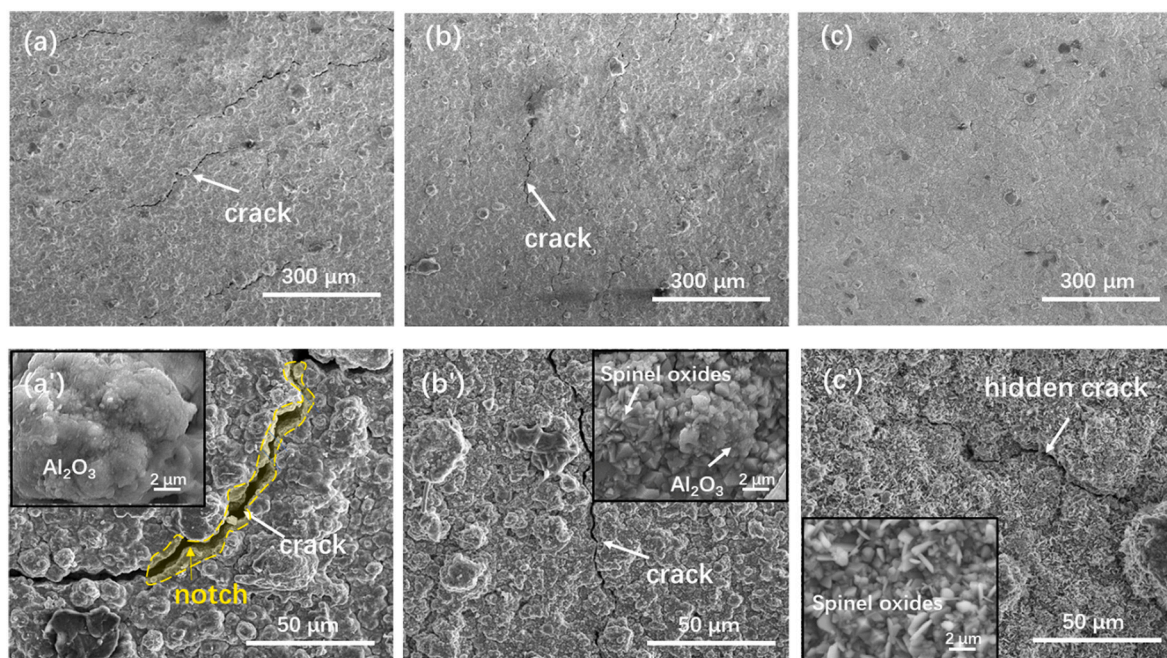


Fig. 6. Surface morphologies of the NiCoCrAlYTa coatings under different bias voltages after thermal shock: (a)(a') -50 V coating, (b)(b') -100 V coating, (c)(c') -150 V coating.

increase in bias voltage.

Fig. 7 shows the cross-sectional SEM images of the coatings after thermal shock. At the low magnification (Fig. 7a–c), it can be observed that many penetrating cracks are existing in -50 V coating and -100 V coating. In contrast, no cracks are observed in -150 V coating. At the high magnification (Fig. 7a'–c'), the width of the cracks in -50 V coating can be up to $3\text{--}4\text{ }\mu\text{m}$, and they almost penetrate the entire coating. As a result, the substrate beneath the crack tip is even subjected to an internal oxidation. Besides, Al_2O_3 scales are formed not only on the coating surface but also around the penetrating cracks, allowing the body of the coating to remain isolated from the external environment. However, by the analysis of EDS mapping for the body region of the coating (Fig. 8), it can be observed that the pores in the coating have been entirely filled by newly formed Al_2O_3 . The porous cauliflower surface morphology of -50 V coating allows the oxygen to rapidly invade into the coating before the intact protective Al_2O_3 scale is formed on the coating surface. Subsequently, the oxygen existing in the pores reacts with the Al in the coating to form Al_2O_3 , ultimately filling the pores. Predictably, these Al_2O_3 may cause additional stress concentration during the thermal shock process due to the difference in the thermal expansion coefficient between coating matrix materials and Al_2O_3 . In that case, more cracks could be formed in the coating, thereby further lowering the thermal shock behaviour of the coating. In the case of -100 V coating (Fig. 7b'), the Al_2O_3 scales on the coating surface and around the cracks also protect the coating body. However, the penetrating cracks are much narrower than those in -50 V coating. Some cracks are even filled with newly formed Al_2O_3 scales. The cracks have not penetrated the entire coating. Instead, they have a tendency to propagate along the horizontal direction. In the case of -150 V coating (Fig. 7c'), the oxide layer exhibits a double-layer structure, where the inner layer is Al_2O_3 scales with about $0.5\text{ }\mu\text{m}$ thickness, and the outer layer is spinel oxides scales with $2\text{--}3\text{ }\mu\text{m}$ thickness. The coating shows a fairly dense structure without any pore or Al_2O_3 existing in the interior of the coating. More importantly, no cracks are formed in the coating. According to the microstructural evaluation of the coatings after thermal shock, it can be clearly known that the thermal shock behaviour of the coating is significantly improved by increasing the substrate bias voltage.

4. Discussion

4.1. Failure mechanism of the coating during the thermal shock process

As mentioned in Fig. 7a', the internal oxidation occurs in the substrate due to the penetrating cracks. Strictly speaking, in that case, the coating loses its protective function for the substrate, although the coating keeps intact without any spallation and internal oxidation. Fig. 9 show the magnified SEM image around the internally oxidised area of the substrate. It can be observed that the penetrating crack does not end at the interface between the substrate and the coating but continuously propagates into the interior of the substrate, ultimately resulting in the internal oxidation of the substrate. When cracks propagates at the interface of coating/substrate, the substrate beneath the crack tip is exposed to the environment, and locally oxidised there. In that case, the formed oxide scales mainly consist of loose spinel oxides due to the relatively low oxidation resistance of the substrate. During the thermal shock process, stress inevitably is generated in the spinel oxide scales. The stress mainly comes from two ways. One is the growing stress of the oxide scales. The spinel oxides possess a high PBR (Pilling-Bedworth ratio) [22], which can cause a huge volume expansion during the growth of the oxides. Here, PBR (Pilling-Bedworth ratio) is the ratio of the oxide molar volume to that of the oxides metal. The other one is the thermal stress caused by the high transient thermal gradient during the thermal shock. Since the spinel oxide scales are limited in the narrow area between the crack tip and substrate, tremendous stress is formed in the oxide scales. This can easily cause the initiation of the crack in the oxide scales. Subsequently, the crack rapidly penetrates through the loose spinel oxide scales, and the substrate is exposed to the environment again. By repeating the above process, the crack can continuously propagate in the substrate. In fact, the above failure model can be considered a kind of stress corrosion (oxidation) behaviour. As well known, stress corrosion usually has a destructive effect on the part. It causes cracks on the part surface, which sharply reduces the fatigue strength of the part. Particularly, for high-speed running parts such as turbine blades, the surface crack is devastating and even leads to the premature scrap of the blade [23,24].

4.2. Effect of the coating microstructure on the thermal shock property

Commonly speaking, the coatings deposited by arc ion plating

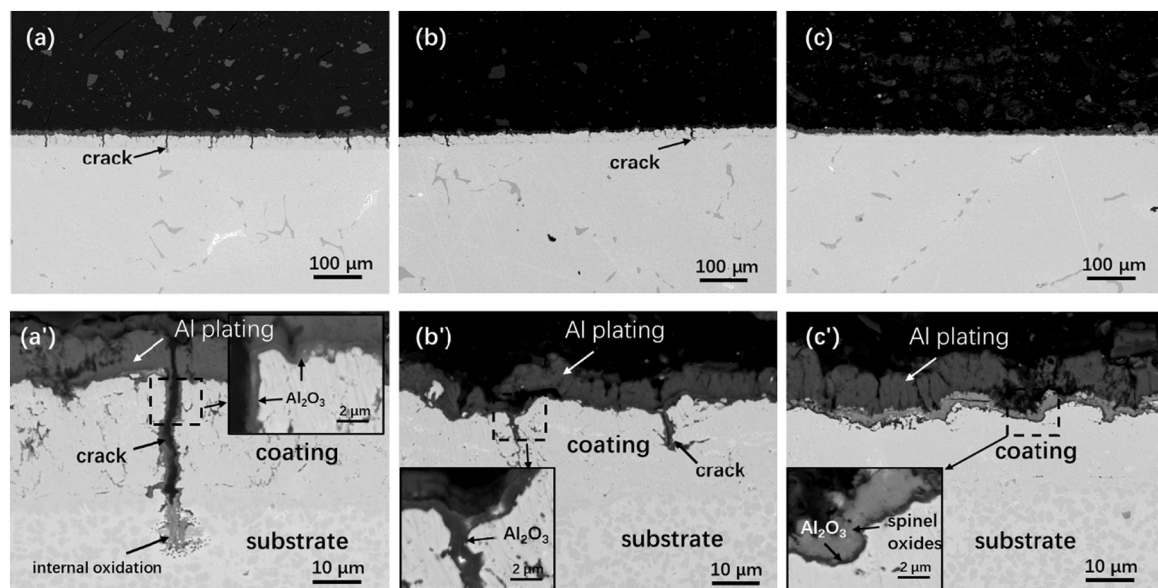


Fig. 7. Cross-sectional morphologies of the NiCoCrAlYTa coatings under different bias voltages after thermal shock: (a)(a') -50 V coating, (b)(b') -100 V coating, (c)(c') -150 V coating.

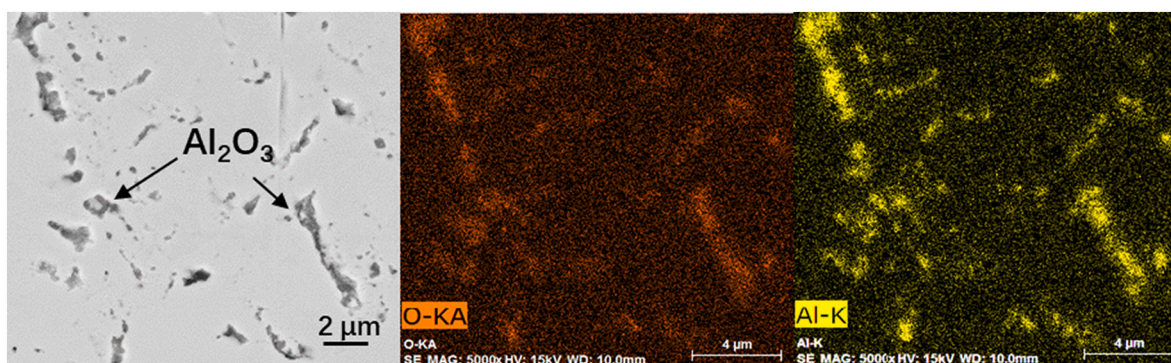


Fig. 8. EDS mappings of the body region in -50 V coating.

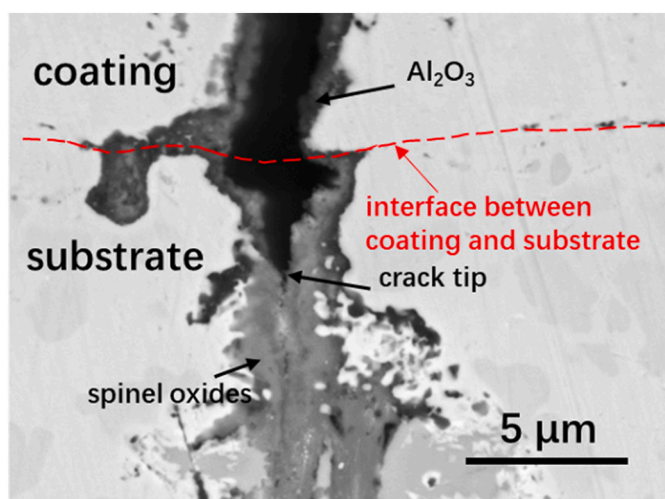


Fig. 9. SEM image of the internal oxidation area in the substrate.

technically show a columnar or a column-like structure like most PVD coatings. The formation of the columnar structure is strongly dependent on the specific deposition mechanism of this technique. Fig. 10 shows the schematic illustrations of the evolution of the coating microstructure during the deposition process and the thermal shock process. The high ionisation degree of arc ion plating makes the vacuum chamber full of metal ions. These metal ions move towards the substrate under the action of the electric field (E) generated by the substrate bias voltage. Under a low substrate bias voltage (-50 V), metal ions possess relatively low kinetic energy. They tend to agglomerate together to decrease surface energy when arriving on the coating surface. Thereafter, the coating is deposited based on the island model (Volmer-Weber), which results in the columnar structural coating. Whereas, under a high substrate bias voltage (-150 V), the metal ions possess relatively high kinetic energy. At the same time, the substrate has a strong absorption to the metal ions. When the absorption effect of the substrate exceeds the agglomeration effect of the metal ions themselves, the coating is built up in the layer model (Frankvan der Merwe), which tends to form a laminar structure at last.

The columnar structure results in a cauliflower-like surface morphology containing a number of notches. During the thermal shock process, the cracks tend to initiate in these notches since the notches more easily cause stress concentration. Besides, the coating is subjected to tensile stress (σ) during the rapid cooling process of the thermal shock test. With the help of the tensile stress, the gap between the column structures provides a favourable channel for the propagation of the crack. As the crack propagates to the substrate, a stress corrosion (oxidation) occurs on the localised substrate beneath the crack tip. It

leads to the continuous propagation of the crack inside the substrate. In contrast, in the case of the coating under a high bias voltage, the notch morphologies on the coating is significantly reduced. On the one hand, the flat surface morphology restrains the initiation of the cracks. On the other hand, the laminar structure also restrains the further propagation of the crack into the interior of the coating. Such a laminar structure is similar to that in the thermal sprayed coating, and the latter generally possesses an ideal thermal shock performance.

According to the above analysis, in order to obtain good thermal shock behaviour, a dense and laminar structure is desirable for MCrAlY coatings. This is exactly contrary to the requirement of the top coating of TBCs. Commonly accepted, due to the low plasticity of the top (ceramic) coating, the top coating is expected to have a columnar structure or vertically segmented cracks in order to release the thermal strain [25,26]. However, this kind of microstructure is obviously not suitable for MCrAlY coatings. As mentioned above, the arc ion plating MCrAlY coatings have a low elastic modulus. The thermal strain can be sufficiently released through the plastic deformation even though the coating possesses a laminar structure.

Although the thermal shock behaviour is significantly improved by increasing the bias voltage, it also reduces the oxidation resistance of the coating at the same time. As shown in Figs. 6 and 7, the number of spinel oxides increases as the bias voltage increases. Commonly accepted, a compact and intact Al_2O_3 scale formed on the coating surface is the premise of high oxidation resistance because the Al_2O_3 scale can effectively isolate the external oxygen and then protect the substrate. When Al in the coating is depleted, the spinel oxides scales are formed. However, they exhibit much less protection than Al_2O_3 scales, eventually lowering the oxidation resistance of the coating. According to the previous research [11], the Al content in -50 V, -100 V and -150 V coating in this study is 5.25 wt%, 4.04 wt%, 2.93 wt%, respectively. For that reason, the oxidation resistance of the coating is decreased as the bias voltage increase. The effect of bias voltage on the composition of the arc ion plating coating has been widely reported by the literature [27–29]. However, the mechanism of this phenomenon is complex and has not been fully clarified yet. Commonly speaking, the loss of Al in the coating under high bias voltage is related to its strong sputtering effect [30] and the low ionisation rate [31].

In order to overcome such contradiction between the thermal shock resistance and the oxidation resistance, a redesign of the target composition is necessary, by which the content of Al in the target should be designed higher than that of the desired coating. To this purpose, the functional relationship between target composition and coating composition should be derived by numerical analysis.

5. Conclusion

In this study, the authors investigated the mechanical and thermal shock properties of the arc ion plating NiCoCrAlYTa coating with different microstructures. Meanwhile, the failure mechanism of the

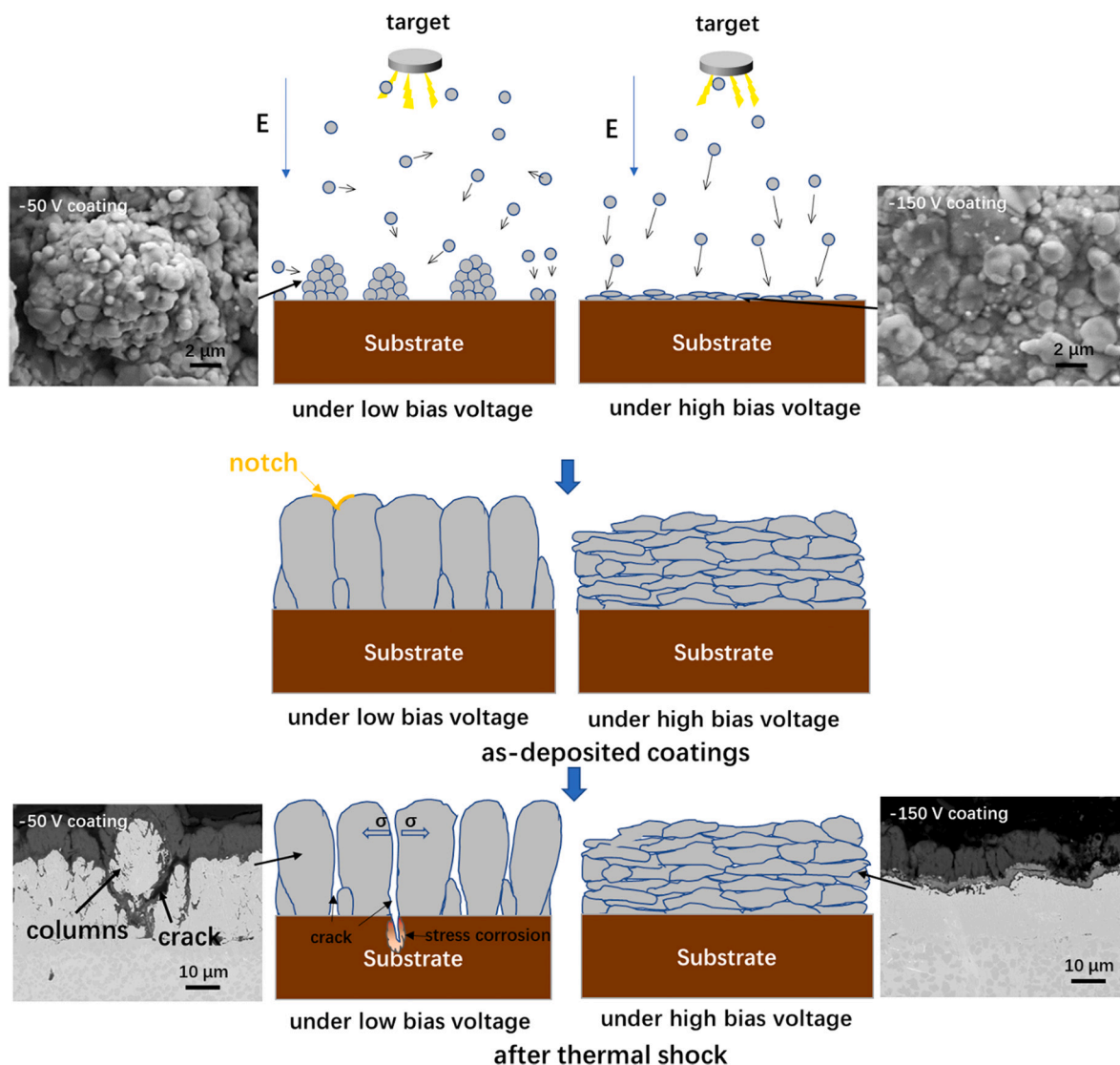


Fig. 10. Schematic illustrations of the evolution of the coating microstructure during the deposition process and the thermal shock process.

coating during thermal shock was discussed as well. The main conclusions drawn are summarised as follows:

1. NiCoCrAlYTa coatings prepared by arc ion plating possess similar hardness but lower elastic modulus compared to those prepared by conventional thermal spraying coatings
2. After the thermal shock, no spallation occurs on the coatings. However, penetrating cracks are formed in the coating with the columnar structure, which ultimately causes stress corrosion in the substrate.
3. The coating with the laminar structure exhibits a better thermal shock property. No cracks are formed in the coating during the thermal shock test. However, in that case, the oxidation resistance of the coating is decreased due to the loss of Al in the coating caused by the increase of the bias voltage.
4. A redesign of the target composition is necessary to overcome the contradiction between the thermal shock behaviour and the oxidation resistance behaviour.

CRediT authorship contribution statement

Shiming Xie: Conceptualisation, Methodology, Investigation, Writing - original draft. **Songshen Lin:** Conceptualisation, Methodology, Investigation, Writing - review & editing. **Qian Shi:**

Conceptualisation, Methodology, Investigation, Writing - review & editing. **Wei Wang:** Methodology. **Chen Song:** Methodology-mechanical properties test, Writing - review & editing. **Wei Xu:** Methodology. **Mingjiang Dai:** Conceptualisation, Supervision, Resources, Writing - review & editing.

Declaration of competing interest

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

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