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# Effect of coating thickness on interfacial adhesion and mechanical properties of Cr-coated zircaloy

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Abstract: The tensile failure behavior of the Cr-coated zircaloy (Zr-4) was investigated. The effect of the coating thickness on the interfacial adhesion of the samples was studied by observing the real-time process of tensile. The interfacial adhesion of the Cr coating was quantitatively evaluated using finite element analysis and shear strength theory. In addition, the crack propagation behavior of the Cr-coated samples was investigated. Results showed that the interfacial shear strength and the interfacial adhesion performance decreased with the increase of the coating thickness. However, the protective effect on the substrate increased with the increase of the coating thickness. Furthermore, the path of the crack propagation at the interface between the coating and the substrate also changed as the coating thickness increased. The cracks at the interface of the thinner coating expanded easily into the substrate, causing the local damage. The cracks of the thicker coating were more likely to propagate along the interface, resulting in the adhesive failure.

Key words: Cr-coated zircaloy; coating thickness; interfacial adhesion; mechanical properties; tensile failure; crack behavior

#### **1** Introduction

Some studies have been devoted to the development of accident-tolerant fuels (ATFs) because of the Japanese Fukushima nuclear accident [1]. An important task of the ATF plan is to develop fuel cladding materials with excellent performance. In particular, the cladding material should be able to maintain the integrity of the structure and perform longer to gain more coping time when a reactor accident occurs [2,3]. Two strategies have been mainly considered for optimized cladding materials. One technique is to develop the other materials, such as SiC composites, iron-based alloys, and multilayer molybdenum, with a better performance, and to replace the zirconium alloys to improve their resistance to

accident [4,5]. The other method is to modify the surface of the Zr-based alloy (usually prepared by a protective coating) to enhance the relevant properties [6,7]. Modifying the surface coating of the Zr-based alloys has been considered as the easiest and the most convenient method which only needs to last for a short time [7]. This technique is used to improve and upgrade the original system of Zr-based alloy cladding. Many materials have been investigated as ATF coating on the surface of the Zr alloy, which include metal coatings (e.g., Cr [8]), ceramic coatings (e.g., MAX phase [9], MAO [10], and CrN [11]), and multilayer composite coatings (e.g., Cr-Zr/Cr/CrN [12] and Mo/FeCrA1 [13]). Among these coatings, the Cr coating has attracted a wide attention due to its excellent comprehensive performance. KIM et al [14] used 3D laser technology to prepare a 90 µm-thick Cr coating and

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found that the coating had excellent adhesion properties at high temperatures. PARK et al [15] developed the Cr coating on a Zr-4 substrate by the arc ion plating technology. The coating exhibited excellent high-temperature oxidation resistance under 1200 °C steam. BISCHOFF et al [16] and BRACHET et al [17,18] prepared a 15  $\mu$ m-thick Cr coating on an M5 substrate by physical vapor deposition. Fretting experiments showed that the coating had good wear resistance. In addition, in the experiment the corrosion of the water chemical environment was simulated in a pressurized water reactor through a static autoclave condition. The results showed that the Cr coating had excellent corrosion resistance under the normal operating conditions.

The important issue for the application of an ATF coating is the improvement of the interface adhesion performance and the failure behavior of the coating. Tensile test [19], scraping test [20], and three-point bend test [21] are widely used to investigate the interfacial bond between the coating and the substrate. Tensile test, which has been widely applied in the interface research in recent years, can provide the uniform deformation of the sample, stably and reliably reflect the inherent properties of the interface, and allow us to observe the experimental process in situ [22]. Combined with the tensile test, AGRAWAL and RAJ [23] developed a new method (A-R model) to evaluate the adhesion performance of the cermet interface [23]. This method has been widely proven to be effective in evaluating the interfacial adhesion between the brittle coatings and the ductile substrates [24]. However, this model is rarely employed in the research of the interface between the Cr coating and the Zr-4 substrate. Current studies have mainly considered the influence of the residual stress and the temperature on the adhesion performance of Cr coatings [25]. However, there are few studies on the optimal range of the chromium coating thickness for the zirconium alloy protection and the interfacial bond strength required for its stability. In addition, it was found that the Zr substrate will be damaged at the Cr-Zr interface in the tensile experiment when the coating is thinner [25], however, without good explanation for this phenomenon. This inspired us to combine theory with experiment to study the influence of the coating thickness on the behavior and the mechanism of the interface failure.

In this work, three coatings with different thicknesses were prepared by multi-arc ion plating technology. Combining real-time monitoring and tensile equipment and A-R theoretical model analysis, the effects of these coatings on the adhesion of the film-based interface were quantitatively evaluated. According to the analysis of the distribution of cracks on the surface and interface of the coatings, the failure behavior and potential mechanisms of the tensile failure of the interface were further analyzed. The effect of the coating thickness on interfacial damage under tensile stress was investigated, and the mechanisms of the crack propagation and the substrate damage at the interface between the coating and the substrate were revealed, which can provide experimental basis and theoretical guidance for the selection of the thickness of the Cr coating on the Zr alloy in nuclear power plants.

#### 2 Experimental

#### 2.1 Material and method

The substrate was Zr-4 alloy, with chemical composition of 1.5% Sn, 0.2% Fe, 0.1% Cr, and balanced Zr (all in mass fraction). The samples were machined into a dog-bone shape and ground with  $240^{\#}$ ,  $400^{\#}$ ,  $800^{\#}$ , and  $1200^{\#}$  grit sandpaper. Then, the samples were cleaned in sequence with ethanol and deionized water in an ultrasonic cleaner, and dried with nitrogen gas.

Multi-arc ion plating technology was used to deposit the Cr coating on the Zr-4 substrate. Two holes (diameter of 1.0 mm) were drilled at both ends of the specimen for the suspension. The samples were rotated at a constant speed (1 r/min) to obtain uniform deposition. The deposition equipment included two high-purity Cr cathodes with parameters of an arc current of 90 A, a negative substrate bias voltage of 120 V, a pressure of 1.2 Pa, and a deposition temperature of  $350 \,^{\circ}$ C. The deposition time was 8, 10, and 12 h, respectively.

The elastic modulus and hardness of the Cr-coated samples were measured by the nanoindentation test. The surfaces of the samples were polished to ensure that the roughness of the samples was less than  $0.05 \,\mu\text{m}$ . Then, the nanoindenter (Hysitron TI 750) was used to measure the elastic modulus and hardness of the samples. The indenter was pressed with a constant strain rate and a maximum depth of 1500 nm. The values of the elastic modulus and hardness of each sample were tested at six different points, and the distance between the adjacent measuring points was longer than 200 nm. Finally, the average value of the measured values from the six points was calculated.

The experimental equipment and sample size are shown in Fig. 1(a). The measurement range of the device was 0-10 kN, and the experiment was performed at a constant speed of 0.05 mm/min under controlled displacement (measured by the microcomputer electronic mechanics tester, ETM104B). A high-magnification optical microscope was equipped for the real-time observation (Cpt-200x, China), and magnification of 200 was used by considering the field of view, clarity, and stability of microscope. The strain of the first crack was determined through repeated three tests, and the scanning electron microscopy (SEM, Apreo 2C, USA) was employed to assist the checking of the strains. The sample was cut from the middle along the loading direction and polished to observe the cross-section after the test. The stress–strain curves of the samples are shown in Fig. 1(b). It can be observed from the figure that the coating significantly enhances the strength and elongation of the zirconium alloy. In addition, the elongation decreases with increasing coating thickness.

### 2.2 Shear strength theory of brittle coating and ductile substrate

The experimental process is shown in Fig. 2. Dog-bone shaped specimens were used in this



**Fig. 1** Experimental equipment and sample size (a) (unit: mm) and stress-strain curves of Samples T1–T3 (with thicknesses of 15, 20 and 25  $\mu$ m, respectively) and substrate (b)



**Fig. 2** Schematic diagram describing shear strength theory of brittle coating and ductile substrate ( $\tau$  is the shear stress, x is distance from Point A,  $\tau_b$  is maximum value of shear stress,  $\lambda_b$  is sine curve wavelength): (a) Tensile specimen; (b) Cross-section diagram of red circle part of specimen; (c) Free body balance; (d, e) Two different distribution forms of shear stress

experiment (Fig. 2(a)). A thin Cr coating was deposited onto a relatively thick substrate, and uniaxial tensile deformation was applied to the sample (Fig. 2(b)). The substrate was deformed by plastic deformation, while the coating was deformed by elastic deformation. The coating was destroyed, and parallel transverse cracks were generated with increasing strain. The relationship between the tensile strength  $\sigma_b$  and the fracture strain  $\varepsilon_f$  of the coating is shown as follows [23]:

$$\sigma_b = \varepsilon_f E$$
 (1)

where *E* is the Young's modulus of the coating. The fracture strain  $\varepsilon_{\rm f}$  changed with the crack density.

As shown in Fig. 2, the distance between the adjacent cracks A and B is L. The deformation of the substrate increased the shear stress at the interface, which was transferred to the coating during the stretching process. AB part was applied to the free body balance condition. Thus, the accumulation of shear stress acting on AB part should be zero when AB part was in a static state at a certain moment. Therefore, the shear stress of the AB part should be antisymmetric, as shown in Fig. 2(d). AGRAWAL and RAJ [23] assumed that the distribution of the shear stress ( $\tau(x)$ ) was in a sinusoidal form as follows:

$$\tau(x) = \tau_{\rm b} \sin\left(\frac{2\pi x}{\lambda_{\rm b}}\right), \ 0 \le x \le \frac{\lambda_{\rm b}}{2}$$
(2a)

$$\tau(x) = 0, \ \frac{\lambda_{\rm b}}{2} < x < \left(L - \frac{\lambda_{\rm b}}{2}\right) \tag{2b}$$

$$\tau(x) = -\tau_{\rm b} \sin \frac{2\pi}{\lambda_{\rm b}} \left[ x - \left( L - \frac{\lambda_{\rm b}}{2} \right) \right], \ \left( L - \frac{\lambda_{\rm b}}{2} \right) \le x \le L$$
(2c)

Then, a part between A and B (the red circled part in Fig. 2(b)) was selected, and this part was also applied to the free body balance condition, as shown in Fig. 2(c). In this free body, the in-plane stress and the shear stress balanced each other. Assuming that the length of this part is x, the inplane stress is a function of distance x [23], that is

$$\sigma(x) = \frac{1}{\delta} \int_{0}^{a} \tau(x) dx$$
(3)

where  $\delta$  is the thickness of the coating.

According to Eq. (3) and the distribution of the shear stress (Fig. 2(d)), the maximum in-plane

stress should appear on the section where  $\tau=0$ , and new cracks should appear in the area of the maximum stress. The stress at the interface will be redistributed when a new crack is formed. This phenomenon indicates the presence of the maximum and minimum crack distances during the tensile process. Based on the analysis of AGRAWAL and RAJ [23], the ultimate goal is to measure the maximum shear stress that can be supported by the interface. Therefore, studying the stress distribution under the maximum crack spacing is necessary. Substituting Eq. (2) into Eq. (3) yields the following equation:

$$\tau_{\rm c} = \frac{\pi \delta \sigma}{\lambda} \tag{4}$$

where  $\tau_c$  is the critical shear stress,  $\lambda$  is the crack spacing at saturation, and  $\sigma = E\varepsilon$ , in which  $\varepsilon$  is the strain when the first crack appears.

Then, through finite element analysis, CHEN et al [26] found that the shear stress at the interface between the coating and the substrate was distributed in an approximate elliptical form (Fig. 2(e)), rather than the assumed sinusoidal form. The formula is as follows:

$$\tau(x) = \frac{\tau_{\rm b}}{\lambda_{\rm b}} \sqrt{\lambda_{\rm b}^2 - \left(x^2 - \lambda_{\rm b}^2\right) + \tau_{\rm b}} \tag{5}$$

The critical shear strength is obtained according to the integral of the shear stress, which can be expressed as

$$\tau_{\rm c} = \frac{1}{\delta} \int_{0}^{a} \tau(x) \mathrm{d}x \tag{6}$$

Finally, CHEN et al [26] revised the form of the A–R model as follows:

$$\tau_{\rm c} = \frac{4\delta\sigma}{\lambda(\pi+4)} \tag{7}$$

#### 2.3 Finite element modeling

In Section 2.2, various distributions of the interface shear stress that led to different equations for calculating shear stress were introduced. Therefore, the finite element analysis was used to determine the distribution of the shear stress at the interface between the Zr alloy and the Cr coating before the calculation (Fig. 3).

The cracks of the coating were periodic in the tensile direction. Therefore, only a part of the unit



Fig. 3 Schematic diagram of boundary conditions and geometric dimensions of finite element model

needed to be considered and studied (Fig. 3(a)). The thickness of the substrate was regarded as infinite compared to the thickness of the coating, thus, only half of the substrate was modeled, as shown in Fig. 3(b). Symmetry constraints about the X axis were set at the bottom of the model, and displacement constraints on the X axis were set on the left side of the substrate. The coating was a free body which was not constrained based on the actual situation. Then, 7% tensile strain was applied to the right side of the substrate. As is shown in Fig. 3(c), the thicknesses of the coating and the substrate were 15 and 650 µm, respectively, and the length of the model was 35 µm. As an elastic material, the Young's modulus and the Poisson's ratio of the Cr coating are 202 GPa and 0.22, respectively. The substrate was defined as an elastoplastic material with Young's modulus of 120 GPa and Poisson's ratio of 0.3. The plasticity data of the substrate were determined from the tensile experiment. Young's modulus was determined from the measurement of nanoindentation, and Poisson's ratio was obtained from Ref. [27]. The simulation was conducted using ABAQUS. The distribution of the interface shear stress was antisymmetric with respect to Y axis, so only half of the length needed to be analyzed.

#### **3** Results and discussion

## 3.1 Microstructure and mechanical properties coating samples

Figure 4 shows the scanning electron micrographs of three coatings with various thicknesses. The morphologies of the surface (Figs. 4(a-c)) showed many spherical particles on the surface of the coating, which were the droplet particles formed by the multi-arc ion plating process [28]. The cross-section morphologies in Figs. 4(d–f) showed that the thicknesses of the coatings were 15, 20, and 25  $\mu$ m, respectively. The coatings had a dense structure, and the interfaces between the coating and the substrate were clearly visible. The microstructure of the samples was obtained by EBSD (Figs. 4(g–i)). The EBSD images showed the columnar crystals of the Cr coating. The coating grains become larger as the coating thickness increases. The three samples were marked as T1 (15  $\mu$ m), T2 (20  $\mu$ m), and T3 (25  $\mu$ m), respectively.

The elastic modulus and the hardness values of the substrate and the Cr-coated samples are shown in Fig. 5(a). The elastic moduli of the substrate, T1, T2, and T3 were (118±3.91), (202.16±8.6), (232.75±10.4), and (188.74±7.2) GPa, respectively, and their corresponding hardness values were (3.18±0.10), (6.79±0.3), (5.57±0.2), and (5.43±0.2) GPa. Figure 5(b) showed the H/E and  $H^3/E^2$  (H is the hardness) ratios. It is well known that the H/E and  $H^3/E^2$  ratios are related to elastic and plastic deformation resistance of the coating [29,30]. The higher H/E ratio indicates a better deformability of the coating. The higher  $H^3/E^2$  ratio indicates the higher resistance to cracks propagation [30,31]. Figure 5(b) shows that the ratios of H/E and  $H^3/E^2$ have a similarly evolutionary tendency with increasing coating thickness. T1 with the 15 µm thickness has the best performance of elastic deformation and resistance to crack initiation and propagation. The H/E and  $H^3/E^2$  of T1 reached 0.0306 and 0.0077 GPa, respectively.

#### 3.2 Tensile failure behavior

Figure 6 shows the evolution of the cracks on the surface of the samples during the tensile process. The strains of the three cracked T1, T2, and T3 were 1.68%, 1.14%, and 0.47%, respectively. Therefore, the corresponding tensile strengths of the three coatings were 3.40, 2.65, and 0.89 GPa according to Eq. (1). These results indicated that the tensile strength of the coating decreased with the increase of the coating thickness. This phenomenon is due to the fact that the thickness of the Cr coating becomes larger and the grain size also increases as the processing time is prolonged. The small size of the grain improves the

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**Fig. 4** Surface (a–c), cross-section (d–f) morphologies, and EBDS images (g–i) of three samples: (a, d, g) T1; (b, e, h) T2; (c, f, i) T3



Fig. 5 Elastic modulus and hardness measured by nanoindentation test (a) and H/E and  $H^3/E^2$  value evolution of Cr coatings with respect to thickness (b)



Fig. 6 Evolution of cracks and surface of samples with different thicknesses during tensile failure process by real-time observation

tensile strength [32]. With the increase of  $\varepsilon$ , the new parallel cracks on the surface of the coating quickly appeared, and the crack density increased rapidly. When  $\varepsilon(T1)$  reached 6.19%, the crack tended to become saturated until it broke. The strains were 2.88% and 1.44% respectively when the cracks of T2 and T3 reached saturation. However, the number of cracks decreased under continuous tension. When  $\varepsilon(T3)=3.56\%$ , obvious cross-linked cracks

parallel to the tensile direction were observed, and the coating began to debond when  $\varepsilon$ =6.15%. Cross-linking cracks occurred due to the Poisson effect [33].

The crack densities of the three samples were evaluated to quantitatively study the surface cracking of the coating under tension. Crack density is defined as the number of cracks per unit length in the tensile direction. The curves of the crack

densities of the three samples obtained from the tensile experiment are shown in Fig. 7. The crack density of T1 increased rapidly before saturation and then remained stable until it was broken. The crack densities of T2 and T3 quickly reached the maximum and then showed a downward trend until saturation. Thus, as shown in previous studies in Refs. [25,27], the thinner the coating was, the greater the crack density was when the coating was saturated. However, the intermediate processes of the crack density were different. The crack density of T2 and T3 decreased dramatically after the strain exceeded a maximum value, and the decrease of T3 was larger than that of T2. These were due to the separation of the interface between T2 and T3, and severer peeling of the coating of T3 (indicated by arrows), resulting in the fact that new cracks become more difficult to form, and the new crack formation rate is lower than the tensile strain rate. The increase of the coating thickness resulted in the change of its adhesion properties. The thicker coating is easy to peel off from the substrate under large strain condition, resulting in the variation of coating mechanical properties. Moreover, the strain in the tensile direction continued to increase, enlarging the crack gap and also resulting in a decrease of the crack density.



Fig. 7 Surface crack density of samples with different thicknesses under tension

#### 3.3 Shear strength of film-substrate interface

Figure 8 shows the distribution of the interface shear stress obtained by the FEM analysis. The small picture at the upper right corner reflects the data obtained by the finite element calculation. The shear stress of the interface is distributed antisymmetrically. Therefore, the data of the other half of the length are completed based on this principle (The black curve at the center of the graph). The red curve is a standard quarter of elliptic curve. The distribution of the interface shear stress between the Zr alloy substrate and the Cr coating is similar to an elliptical curve. This result is consistent with that proposed by YANAKA et al [34] and JEONG and KWON [35]. Part of the interface shear stress was close to zero (blue arrow area), and this part corresponded to the area with the largest in-plane stress in the A-R model. LI et al [30] used FEM to make a model with cracks and found that as the width of crack became smaller, the data were closer to an elliptical curve. Figure 8 shows that Eq. (7) should be considered to calculate the shear stress of the Zr-Cr interface.



Fig. 8 Interfacial shear stress distribution

Figure 9 shows the cracks on the surface of the sample at the tensile saturation stage. As the coating thickness increased, the crack interval  $\lambda$  increased. A 2000 µm line was drawn in the figure, and the number of the cracks was counted. The average crack interval was determined through dividing 2000 by the corresponding number of the cracks (N), i.e.,  $\lambda_{\text{average}} = 2000/N$  the shear strength of the interface was calculated by Eq. (7). The data and calculation results are listed in Table 1. The average shear strengths of the interfaces of T1, T2, and T3 were 342.50, 193.24, and 68.33 MPa, respectively. As the coating thickness increased, the shear strength of the Zr-Cr interface decreased. The shear strength of the interface indicated the ability of the interface to resist damage. Thus, the thinner the coating was, the stronger the resistance to shear failure was at the interface between the coating and the substrate during the tensile test. In addition, this

result indicated that the thinner the thickness was, the better its adhesion performance of the coating was.

### **3.4** Failure behavior of samples with different coating thicknesses

Figure 10 shows the morphology of the sample surface after tensile fracture. Representative areas in the fracture and nonfracture regions were characterized and analyzed. The fractures of the three samples showed the typical morphology of the tensile strain, which fractured along  $45^{\circ}$  due to the maximum stress in the direction of  $45^{\circ}$ . The coating of T1 adhered tightly to the substrate after breakage,

showing that the coating had good adhesion performance. However, the coatings of T2 and T3 were debonded, and as the thickness of the coating increased, the degree of debonding became severer. Part of the coating debonding indicated that not only the interface between the coating and the substrate became separated, but also the cracks formed parallel to the tensile direction inside the coating, which caused the breakage of the coating. The thicker the coating was, the easier the production of cross-linked cracks was.

The surface morphologies of the samples were further analyzed by SEM (Fig. 11). Many spherical particles were found on the surface of the coating,



Fig. 9 Surface cracks at tensile saturation stage: (a) T1; (b) T2; (c) T3



**Fig. 10** Surface morphologies of samples after fracture: (a-c) Fracture of T1, T2 and T3, respectively; (d-f) Non-fracture of T1, T2 and T3, respectively

Coating thickness, $\delta/\mu m$	First crack strain/%	Elastic modulus/GPa	$\lambda_{min}/\mu m$	$\tau_{\rm max}/{ m MPa}$	$\lambda_{max}/\mu m$	$\tau_{\rm min}/{ m MPa}$	$\lambda_{average}/\mu m$	$ au_{\mathrm{average}}/\mathrm{MPa}$
15(T1)	1.68	202.16	32	891.88	113	252.57	83.33	342.50
20(T2)	1.14	232.75	50	594.59	268	110.93	153.85	193.24
25(T3)	0.47	188.74	89	139.60	278	44.69	181.82	68.33

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Fig. 11 Surface morphologies of samples with different coating thicknesses after fracture: (a-c) T1; (d-f) T2; (g-i) T3

which were the droplet particles caused by the multi-arc ion plating [28]. These droplet particles were generated by the arc point emission on the Cr target and were usually considered as a technology defect because they would reduce the coating performance [36,37]. Some cavities were observed on the surface of the coating (Fig. 11(a)), and these cavities were caused by the falling of the droplet particles. Larger droplet particles would not fall from the surface and would cause crack deflection (Figs. 11(c) and (g)). The parallel cracks on the surface of the coating were not continuous but were composed of many small cracks as determined after magnified observation (Figs. 11(b) and (d)). Several defects in the coating produced cracks at the same

time during deformation, and each defect began to expand and finally converged to form large parallel cracks. As the thickness of the coating increased, cross-linked cracks gradually appeared on the surface of the coating (Figs. 11(e), (f), (h), and (i)). The cross-linked cracks were formed due to the Poisson effect. The substrate underwent strong plastic deformation during tensile strain, while the deformation of the coating was considerably small, which was incompatible with the deformation of the substrate. Therefore, perpendicular to the tensile direction, the coating underwent a compression effect and was bent due to the deformation incompatibility, resulting in cross-linked cracks parallel to the tensile direction [33]. The crosslinked cracks were more likely to appear when the plastic deformation of the sample was larger. The Poisson effect was also confirmed according to Fig. 10, which showed that more cross-linked cracks appeared near the tensile fracture. Moreover, Figs. 10 and 11 show that the thinner the coating was, the better the bending resistance was. Figure 11(i) displays the cross-section of the cross-linked crack (small picture at the upper right corner). The cross-section of the Cr coating showed intergranular and transgranular fracture.

Figure 12 shows cross-sectional morphologies of the fractured samples. Comparison of the cross-sections of the three samples showed that as the coating thickness increased, the interfacial adhesion performance declined. The coating of T1

remained completely adhered to the substrate after tensile fracture, which indicates that the coating has excellent interfacial adhesion properties (Fig. 12(a)). However, the interface of T2 showed a certain degree of damage (Fig. 12(d)), and T3 showed a greater degree of damage (Figs. 12(g) and (h)). These results are consistent with those of the interface shear strength in Section 3.2. The greater the shear strength was, the better the interface damage resistance was, and the less likely the formation of transversal cracks was. Many microcracks were observed at the interface (Figs. 12(c), (e) and (g)), indicating that the cracks initiate at the interface, which is consistent with the results of previous studies [27]. In addition, analyzing the failure behavior of coating is helpful



**Fig. 12** Cross-sectional morphologies of samples with different coating thicknesses after fracture: (a–c) T1; (d–f) T2; (g–i) T3

for understanding the effect of coating on the substrate at the interface. Coating is applied to better protecting the substrate; however, we found in some of our previous studies that the substrate of the coated sample was damaged because of the environmental factors. Therefore, the status of the substrate at the Zr-Cr interface was investigated in this work. Tear-like failure of the substrate was observed at the interface of T1 (Fig. 12(b)). This phenomenon was consistent with the study of JIANG et al [25,27]. However, this kind of tearing failure was only found in the thinner coating samples. With the increase of the coating thickness, no such a tearing failure was observed on the substrate. Comparison of the three samples (Figs. 12(c), (f), and (i)) showed that the thinner the coating was, the greater the degree of crack intrusion into the substrate during tensile deformation was. Thus, the thinner the coating was, the better the adhesion performance was; however, the easier the generation of damage to the substrate during tension was. The adhesion performance and protective effect of the coating were two contradictory factors here.

Studies have shown that the fracture mode of the Cr-Zr system is very similar to that of the brittle coating-ductile substrate system [38]. During the tensile process, the shear stress generated at the interface was moved to the coating to form cracks due to strain incompatibility [39]. Therefore, cracks generally start from the interface (Fig. 12(e)). JIANG et al [27] used finite element calculations to prove that the vertical crack tip is a high-stress concentration area; that is, the stress concentration area occurs at the interface, which leads to the plastic failure of the substrate at the interface under large tension. Interestingly, this phenomenon has two different fracture modes (Fig. 13(a)). As the coating thickness increased, this phenomenon gradually disappeared. In response to this interesting phenomenon, we tried to analyze its mechanism, as shown in Fig. 13. Contrary to the theoretical horizontal interface, the real interface was uneven and undulating (Fig. 13(b)), similar to a wave. When the crack appeared in the wave valley, the stress was concentrated in the valley, as shown in Fig. 13(c). When the crack appeared at the wave peak, the crack propagated along the slope to the valley, resulting in an angular deviation of the stress at the interface, as shown in Fig. 13(d). The continuous shear stress at the interface acts as a



**Fig. 13** Interface failure mechanism of samples with different coating thicknesses

driving force to promote the formation of interface cracks [40]. When the stress and the energy release rates reach a critical value, cracks will appear. However, as the coating thickness increases, the interface shear strength decreases, and the interface fracture toughness decreases [22]. These characteristics indicate the ability of the interface to resist damage decrease [41]. Therefore, when the stress is concentrated at the interface and the energy needs to be released, the samples with thicker coatings will be more likely to form interface cracks. This phenomenon shows that the energy released along the interface causes the cracks to expand along the interface. By contrast, the energy of the thinner coated samples at the interface is not easy to be released along the interface. Instead, this energy diffuses into the substrate to cause damage. In addition, as previously mentioned, the cracks propagating to the substrate form different expansion angles because they are in different specific positions. As shown in Fig. 13(c), when the stress concentration was at the valley, the crack vertically expanded into the substrate. When the stress concentration was at the peak of the wave, the crack entered the substrate at a specific angle along the slope (Fig. 13(d)).

In engineering applications, Zr-alloy cladding coating is applied with the assumption that the coating cannot affect the performance of the substrate. However, the crack behavior on the interface of samples with different coating 2684

thicknesses showed an interesting phenomenon. Thinner coatings have excellent adhesion properties but are more likely to cause damage to the substrate. After the thickness of the coating reached a certain value, it acted as a sacrificial layer and protected the substrate from being affected. Hence, the adhesion performance of the coating and the protective effect on the substrate became two contradictory factors. Thus, choosing a suitable thickness is significant to ensure excellent adhesion performance under actual working conditions and minimize the effect on the substrate. In addition, the actual working temperature of the cladding tube was 320 °C. A previous study has shown that the ductile-to-brittle transition temperature (BDTT) of the Cr coating is 150-500 °C [32]; that is, the actual working temperature of the coating is just within its BDTT range and will also affect the choice of coating thickness.

#### **4** Conclusions

(1) During the tensile test, the Cr coating was brittle at room temperature. The crack density of the thinner coating sample increased rapidly and became saturated in a short time. With the increase of the coating thickness, the crack density of the sample increased and then decreased to saturation. In addition, the greater the thickness of the coating is, the lower the tensile strength and saturation crack density are caused.

(2) The finite element analysis showed that the distribution of the interfacial shear stress in Cr–Zr system under tension was approximately elliptical. The interfacial shear strengths of the samples with different coating thicknesses were calculated using the shear strength theory. As the coating thickness increased, the interfacial shear strength decreased.

(3) Under tension, due to the deformation incompatibility between the coating and the substrate, the Poisson effect of the coating caused the bending deformation to be perpendicular to the stretching direction. As the thickness of the coating became larger, the preparation time was prolonged, the grain size of the coating was enlarged, and the bending resistance decreased. These characteristics ensure the surface of the coating more conducive to form cross-linked cracks. Therefore, the thicker the coating is, the severer the damage is.

(4) The greater the thickness of the coating is,

the easier the propagation of cracks along the interface is, which makes the coating debond. However, the cracks at the interface of the sample with thinner coating easily extend to the inside of the substrate during tension, which damages the substrate. As the coating thickness is reduced, the interfacial adhesion performed better, whereas the substrate is easier to damage.

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### 涂层厚度对镀铬锆合金界面结合力和力学性能的影响

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**摘 要:**研究镀铬锆合金(Zr-4)的拉伸失效行为。通过对拉伸过程的实时观察,研究涂层厚度对界面附着力的影响。使用有限元分析和剪切强度理论定量评估 Cr 涂层的界面附着力。此外,还研究镀铬样品的裂纹扩展行为。 结果表明,随着涂层厚度的增加,界面剪切强度和界面黏附性能下降。然而,对基材的保护作用随着涂层厚度的 增加而增加。此外,涂层与基体界面处裂纹扩展的路径也随着涂层厚度的增加而变化。较薄涂层界面处的裂纹很 容易扩展到基体中,造成局部损伤。较厚涂层的裂纹更容易沿界面扩展,导致黏合剂失效。 关键词: 镀铬锆合金;涂层厚度;界面黏附;力学性能;拉伸失效;裂纹行为

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