Numerical modeling of cracking behavior in Cr coating for ATF cladding under three-point bending

Mingduo Yuan\textsuperscript{1}, Ziyun Pan\textsuperscript{1}, Zhenyu Zou\textsuperscript{1}, Weijian Zhang\textsuperscript{1}, Mingyue Du\textsuperscript{1}, Jishen Jiang\textsuperscript{*}, Xianfeng Ma\textsuperscript{*}

\textsuperscript{1}Sino-French Institute of Nuclear Engineering and Technology, Sun Yat-Sen University, Zhuhai 519082, Guangdong, China
\textsuperscript{*}Corresponding Authors email: jiangjsh3@mail.sysu.edu.cn (Jiang J); maxf6@mail.sysu.edu.cn (Ma X)

Abstract: In-situ three-point bending tests and finite element modeling based on the cohesive zone model were developed to study the stress evolution and cracking behavior of the Cr coated Zr-4 alloy for accident tolerant fuel claddings. The initiation and propagation of micro-cracks were captured by in-situ observation and predicted by the numerical simulation. The results showed that vertical cracks first initiated from the coating surface and propagated to the Cr/Zr\textsubscript{4} interface. Under larger bending strain, interfacial cracks began to initiate from the vertical crack tips driven by large local stress concentration.

1. Introduction
After the Fukushima nuclear accident, great attention has been focused on developing the accident tolerant fuel (ATF) materials to improve the accident resistance of the nuclear materials under both design-basis accident conditions and beyond-design-basis conditions [1-3]. Surface coatings on the fuel claddings were regarded as one of the most favorable methods. Among different kinds of ATF coatings, Cr coating was proved to be one of the optimal consideration due to its superior oxidation and corrosion resistance, irradiation resistance, etc. [4-6]. However, the mechanical properties of the Cr coated-Zr cladding system were seldom investigated, although they played a crucial role in the application of the Cr coatings.

The mechanical properties of the Cr coated Zr\textsubscript{4} substrates are evidently related to the cracking behavior, including both surface cracking and interfacial cracking of the Cr coating [7]. Under the external loading, vertical cracks may appear in the Cr coating. Under the continuous loading, these cracks will propagate into the substrate or deflect to the interface to form interfacial cracks. The crack path is high dependent on the fracture toughness of the coating and the interfacial strength of the coating-substrate. When the interfacial adhesion is weak, interfacial cracks will be formed from the vertical crack tips driven by local stress concentration. When the interfacial adhesion is strong enough, vertical cracks in the Cr coating will penetrate into the substrate, leading to the failure of the coated sample [7]. Therefore, the cracking behavior can reflect the mechanical properties of the coated sample. In our previous work [8,9], evolution of surface cracks were successfully captured by in-situ tensile tests. However, interfacial cracks might be initiated under large deformation. The initiation and propagation of interfacial cracks and the competition between the interfacial and vertical cracks were unclear. Thus, the effective experimental method and numerical model should be developed to study the cracking behavior of the ATF coatings.
In this work, in-situ three-point bending tests were developed to capture the cracking behavior of the Cr coated Zr-4 substrate. A finite element model (FEM) based on the cohesive zone model (CZM) was built to study the stress evolution and crack propagation of the Cr coating. The cracking modes of the Cr coating samples were further investigated.

2. Materials, experimental test, and finite element modeling

The Zr-4 substrate was machined into cuboid three-point bending samples, the geometry of which was shown in figure 1(a). After polishing and cleaning processes, Cr coatings were deposited on the Zr-4 substrate by magnetron sputtering technique. As shown in figure 1(b), the thickness of the as-deposited Cr coating was about 15μm, and no original micro-voids or micro-cracks appeared.

![Figure 1](image_url)

Figure 1. (a) Geometry of the three-point bending sample, unit: mm, (b) cross-sectional morphology of the as-deposited Cr coated Zr-4 substrate, (c) In-situ mechanical test setup equipped with a high-magnification optical microscope, (d) view of the mechanical loading system.

As shown in Figure 1(c), in-situ three-point bending tests were carried out at room temperature on a mechanical testing system equipped with a high magnification optical microscope. The sample was fixed in the loading system by applying small preloading, as shown in Figure 1(d). During the bending test, the micro-deformation and crack evolution of the Cr coating-Zr4 substrate could be observed by the optical telescope in real time. Three-point bending tests were performed under a displacement-control mode at a constant rate of 5×10^{-3} mm/s. The bending test was suspended for several times to capture the images of the cracking in the coated sample.
To model the stress and crack evolution of the Cr coated sample, a two-dimensional FE model was built in ABAQUS, and the geometry and meshes were shown in Figure 2. The geometry was the same as the test sample, and the meshes in the middle region near the coating were refined to obtain enough calculating precision. The Cr coating was regarded as elastic material and the substrate was regarded as elasto-plastic material. The mechanical parameters could be found in Ref. [8,10]. To model the vertical crack in the Cr coating and the interfacial crack between the coating and the substrate, the cohesive elements in zero thickness were inserted in the FE models, as shown in figure 2. The damage-based CZM assumes that once the cohesive element reaches a critical value, damage will occur and accumulate under continuous loading. Once damage value reached one, crack will be initiated. Detailed description could be found in Ref. [8,10]. In the CZM, the fracture strength $\sigma_0$ and the fracture toughness $G_0$ for both the Cr coating and the interface should be determined first. Due to lack of published data, parameter sensitivity analyses were carried out, and finally for the Cr coating, $\sigma_0$ and $G_0$ were set as 150 MPa and 200 J/m$^2$, respectively, and for the interface, $\sigma_0$ and $G_0$ were set as 150 MPa and 100 J/m$^2$, respectively.

3. Results and discussion

3.1. In-situ observation

Figure 3 shows the cracking behavior of the Cr coated Zr-4 substrate under three-point bending based on in-situ observation. Vertical cracks were first generated in the Cr coating driven by local tensile stress. As the loading continued, vertical crack density increased and finally entered a plateau stage, which is consistent with the in-situ tensile test [8]. It is worth noting that at large deflection, interfacial cracks began to initiate from the vertical crack tips, which were driven by the large local interfacial peeling stress and shear stress. However, even under severe deformation, spallation of the Cr coating did not occur, which reflects the good interfacial adhesion of the coating.
Figure 3. In-situ observation on the cracking behavior the Cr coated Zr-4 substrate under different deflection, d: (a) d=0 mm, (b) d=0.6 mm, (c) d=0.9 mm, (d) d=1.3 mm.

3.2. Finite element results
As shown in the experimental results, both vertical and interfacial cracks appeared under bending. The initiation and propagation of the cracks were believed to be driven by the local stresses. Figure 4 displays the stress and vertical crack evolution based on the FE calculation. It is shown that damage appeared in the coating layer when the deflection, \( d \), reached 0.04 mm. The damage led to local stress degradation in the Cr coating. When \( d \) reached 0.260 mm, vertical crack began to initiate from the coating surface and then propagate vertically to the interface. Note that the stress around the crack released greatly. Namely, the presence of crack leads to stress redistribution. Under continuous loading, the crack opened greatly and the stress concentration in front of the crack tip became more significant. The large stress in the substrate beneath the coating might lead to crack penetration into the substrate.

Besides, the interfacial crack initiated from the crack tip was also modeled. The FE results shown in Figure 5 (a) is consistent with the experimental result shown in Figure 5(b). The initiation of the interfacial crack greatly released the local stresses around the vertical crack tip. As the bending loading increased, the crack began to propagate along the interface. Based on the experimental and FE results, the cracking behavior followed a similar trend in those typical brittle coating-ductile substrate system under external loadings. The brittleness of the Cr coating might be related to the microstructure and the large residual stress generated during deposition process. Once the microstructure of the Cr coating is optimized and the residual stress is eliminated by some special deposition process and heat-treatments, the mechanical properties of the Cr coating would be improved and different fracture modes would be expected.
4. Conclusions

In-situ three-point bending tests and finite element simulation based on the cohesive zone model were developed to study the stress evolution and cracking behavior of the Cr coated Zr-4 substrate for accident tolerant fuel claddings. The initiation and propagation of both vertical and interfacial cracks were captured by in-situ observations and predicted by the finite element calculation. The results showed that vertical cracks were first generated in the coating and then propagated to the interface.
Under larger bending strain, interfacial cracks began to be formed from the vertical crack tips driven by large local stress concentration. However, even under severe deformation, spallation of the Cr coating is still not observed, which reflects good interfacial adhesion of the present Cr coating.

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