# SCRATCH AND FRETTING WEAR CHARACTERISTICS OF SURFACE MODIFIED CLADDINGS FOR ACCIDENT-TOLERANT FUEL

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## ABSTRACT

Tribological behaviours of CrAl-coated fuel claddings for accident-tolerant fuel (ATF) have been examined to verify the reliability of the coating layer, focusing on not only the deformation behaviour of the coating layer under a scratching test, but also the wear mechanisms including material transfer under the fretting wear conditions. Recent studies indicate that the protective layer of Zr cladding coated by outstanding corrosion-resistant alloys is considered as one of the key factors for enhancing the accident tolerance under (beyond) design-based accidents. Regardless of their outstanding corrosion resistance in high temperature steam, however, mechanical damage such as grid-to-rod fretting, debris fretting, cracking by high frequency fatigue, and coating failure by mechanical contact should be examined because CrAl-coated layers should be maintained without any significant degradation during normal operation. In this study, four kinds of CrAl-coated claddings with a different negative substrate bias were prepared with an arc ion plating method. After the scratch tests, no gross spallation or tensile cracks of the coating layer were found in the entire stroke length up to a normal load of 50 N. Fretting wear results show that a significant wear resistance of CrAl-coated claddings can be achieved compared with Zr-based cladding. Owing to the relatively high bulk hardness of CrAl-coated claddings, the material transfer and adhesive wear were dominant wear mechanisms against a Zr-based grid spring. Based on the test results, the effects of negative substrate bias on the tribological behaviours of CrAl-coated fuel claddings were examined by focusing on the reliability of the coating layer during normal operation.

KEYWORDS: Accident-tolerant fuel, surface modified cladding, scratch, fretting wear, Zirconium alloy, spacer grid.

## 1. Introduction

As one of the strong candidates for accident-tolerant fuel (ATF) cladding, Zr-based claddings coated with various corrosion resistance alloys have being developed to increase the high temperature oxidation resistance and mechanical strength under normal operation and a (beyond) design-basis accident [1-4]. Various coating methods including arc ion plating (AIP), a cold spray, and laser beam scanning, were applied to form protective layers on the outer surface of the Zr-based claddings. For the successful loading of ATF cladding without any significant design changes of the fuel assembly parts, it is important to maintain these coating layers without excess mechanical and corrosion damages during normal operation. Consequently, these protective coating layers should be preserved during normal operation to achieve the intrinsic roles of ATF cladding under severe accident conditions. Previous results [1,3,5] indicate that surface-modified Zr-based claddings with corrosion resistant alloys showed a decrease in the corrosion rate by at least two orders of magnitude under normal operation and simulated severe accident conditions. However, the evaluation results of the mechanical damage such as grid-to-rod fretting, debris fretting, and crack formation by high frequency fatigue under flow-induced vibration (FIV) are still insufficient. In this work, CrAl-coatings deposited by an AIP method at different negative substrate bias were prepared and submitted to a scratch test and fretting wear test. The objectives of this study are to examine the deformation behaviour and adhesion strength of the coating layer using a constant load scratch test in room-temperature unlubricated conditions, and to compare the fretting wear behaviours of CrAl-coated claddings against a current Zr-based spacer grid in room-temperature water.

(a) Scratch tester (b) Fretting wear tester

Fig 1. Schematic views of scratch and fretting wear tester for evaluating the mechanical properties of CrAl coating layer

## 2. Experiments

### 2.1 CrAl-coated cladding and spacer grid specimen

A Zirconium fuel cladding with dimensions of 9.5 mm in outer diameter and 400 mm in length was used as the substrate material. The nominal composition of the target used to prepare the CrAl coatings was Cr-15Al in weight percent. An AIP machine was employed to deposit the CrAl coatings on Zr cladding samples. To obtain CrAl coatings with optimized performance under high-temperature corrosion resistance, the coatings were deposited under different bias voltages ranging from -100 to -250 V (i.e., -100, -130, -150 and -250V) with the same arc current of 60 A, and working pressure of 20 mTorr. Coated claddings were ground on 1200# SiC paper to obtain similar roughness conditions with a commercial Zr fuel cladding. After that, a low-speed cutter was used to cut the coated cladding into samples of 12.5 mm in length for scratch and fretting wear tests. Four kinds of CrAl-coated cladding were fabricated with different negative substrate bias, as listed in Table 1. In the fretting wear tests, a Zr-based grid spring with a concave shape was used as a spacer grid, which was intended to wrap around the fuel cladding in the circumferential direction. Other detailed characteristics of the spacer grid can be found in a previous study [6].

| Label | Bias voltage | Post-polishing | Roughness | Mean Coating thickness |
|---|---|---|---|---|
| A-100V | -100 V | Yes | 0.127 μm | ~ 10 μm |
| B-130V | -130 V | Yes | 0.292 μm | ~ 50 μm |
| C-150V | -150 V | Yes | 0.212 μm | ~ 50 μm |
| D-250V | -250 V | Yes | 0.324 μm | ~ 50 μm |
| Zr | N/A | As-received | ~0.3 μm | N/A |

Tab 1: Summary of CrAl-coated cladding samples and uncoated Zr cladding

### 2.2 Scratch and fretting wear tests

A constant load scratch tester to evaluate the deformation behaviour of the CrAl coating layer was specially designed, as shown in Fig. 1a. A normal force can be applied from 0 to 50 N by adjusting the dead weights. In this study, three kinds of scratching speed (i.e., 0.1, 0.5, and 5 mm/s) were applied with an initial normal force of 2 N and stroke length of 5 mm under room-temperature unlubricated conditions. A fretting wear tester used in this study is described in Fig. 1b. The detailed test procedure including the installation methods of the cladding and grid specimen, and the specimen array, can be found in our previous work [7]. A peak-to-peak amplitude of 100 μm was applied up to fretting cycles of $1x10^6$ with the same initial normal force of 10 N and frequency of 30 Hz in room-temperature water. After the fretting tests, the worn area of the CrAl coating layers was observed using an SEM and optical microscope for analysing and measuring the wear mechanism and characteristics of each worn area, respectively. Next, the wear volume and depth profile of the CrAl coating layers were measured using a 3-D surface profilometer, and their results were compared for each test condition.

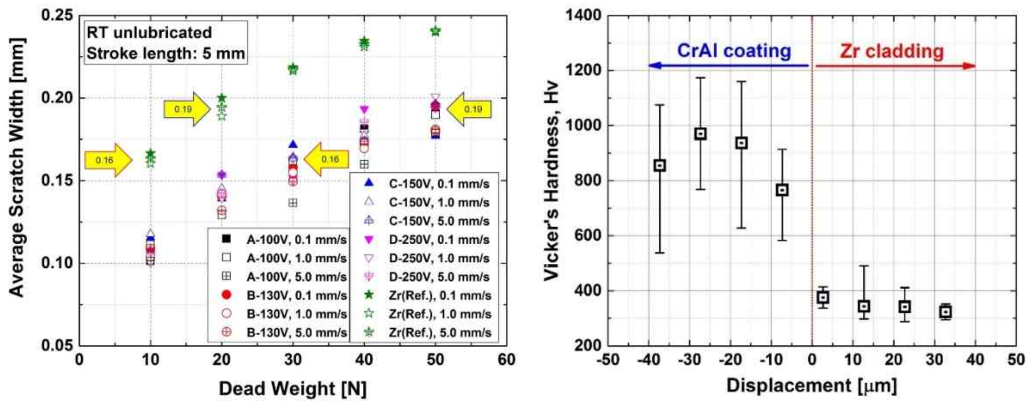
### 2.3 Nanoindentation test and measurement

The characteristics of the coating layer were evaluated using a computer-controlled nanoindentation tester (CSM instruments). The indenter was controlled at a constant loading/unloading rate of 60 mN/m until the maximum load of approximately 30 mN was reached and then held about 10 s. From the results of loading-unloading curves, hardness and elastic modulus of both CrAl coating layer and Zr-based cladding are calculated based on the Oliver and Pharr method [8]. After the scratch and fretting wear tests, the damaged area of the CrAl coating layers were characterized using an optical measurement microscope and scanning electron microscopy (SEM). SEM images of the entire scratch track were observed to examine the formation of cracks and failure behaviour for evaluating a critical load of coating-substrate adhesion and scratch hardness of the CrAl coating layer. In addition, the wear volume and depth profile of the contact area were measured using a 3-D surface profilometer, and their results were compared for each test condition.

## 3. Results and discussion

### 3.1 Deformation behaviours of CrAl coating layer

A constant load scratch test and nanoindentation measurement were conducted to evaluate the deformation behaviours and mechanical properties of different CrAl coatings. Fig. 2a plots the measurement results of the average scratch width ($W_{avg}$) as a function of the dead weight. The value of $W_{avg}$ gradually increases with the applied dead weight because of the contact area expansion. In general, the wider the scratch width was, the lower the bulk hardness of the coating layer [9-10]. In addition, the hardness of the coating layers increases with an increase in negative substrate bias. This is because a higher negative substrate bias indicates more intense ion bombardment, and finally densification of the coatings increase their bulk hardness. Under the same dead weight, however, the $W_{avg}$ of various CrAl coatings show a relatively weak dependency of the applied negative substrate bias. This result can be explained in that each CrAl coating layer used in this study shows a different roughness and coating thickness by surface polishing treatment after the AIP coating processes, which can change the initial deformation behaviours of the constant load scratch tests. In addition, a negative substrate bias effect could be eliminated by removing the outer coating layer for controlling the coating thickness and its roughness.




(a) Scratch width (b) Nanoindentation results

Fig 2. (a) Measurement results of average scratch depth ($W_{avg}$) with dead weight at various CrAl-coated claddings and uncoated Zr cladding, and (b) typical example of hardness variation at CrAl coating layer (D-250V)

Fig. 2b shows a typical example of nanoindentation results at the CrAl coating layer and it is apparent that the mechanical properties of the coating layer are continuously changed with the deposition direction. Therefore, after polishing treatment, the final thickness and surface roughness are considered as important factors determining the mechanical behaviours of the CrAl outer coating layer rather than the applied negative substrate bias itself. This result indicates that if it is difficult to obtain uniform mechanical properties of the CrAl coating layer, the thickness reduction removed by the outer surface polishing treatment should be determined for considering the optimized mechanical properties of the CrAl coating layer.

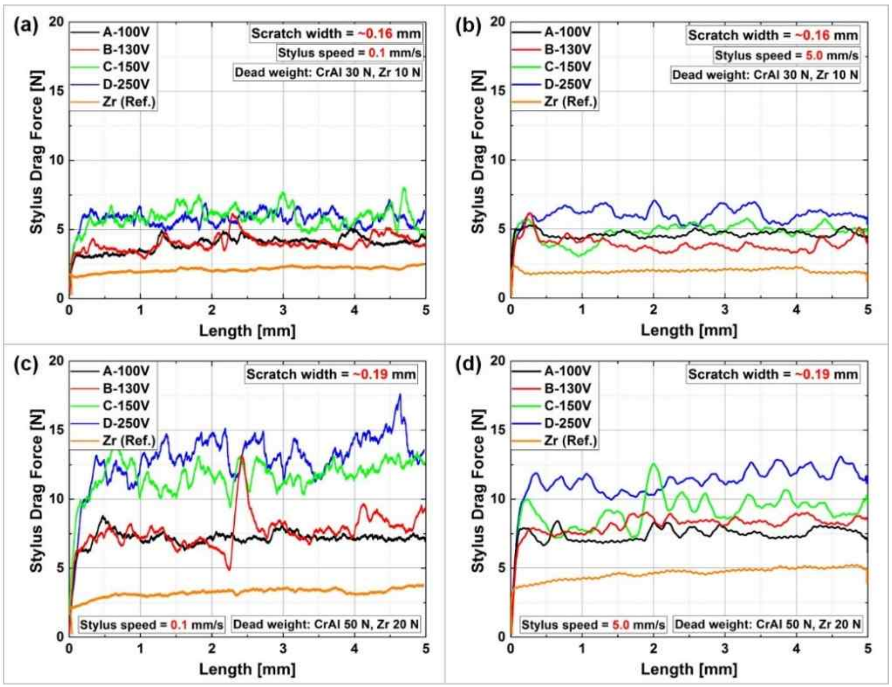
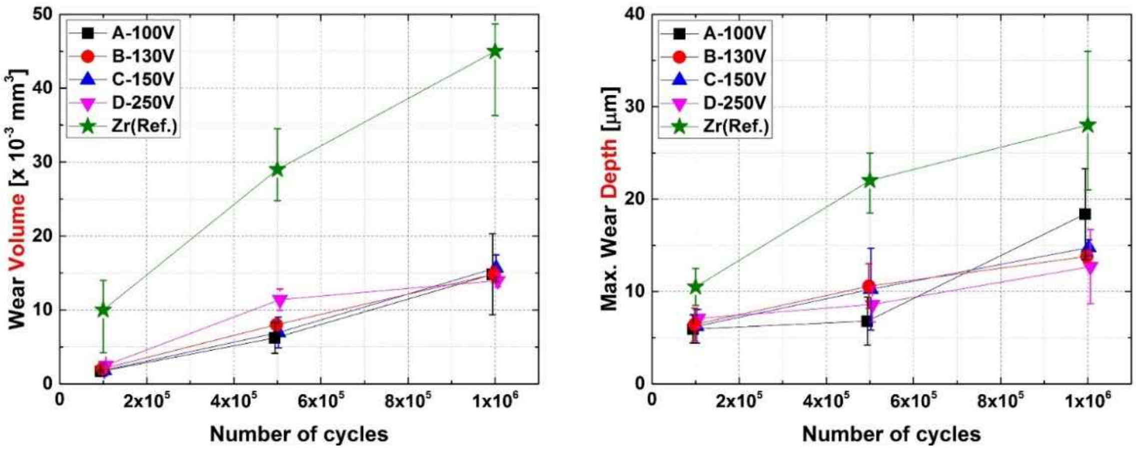



Fig 3. Variation of drag force at similar scratch width region.

For a more accurate comparison of the surface hardening behaviours with a negative substrate bias, a stylus drag force that determines the amount of plastic deformation under scratch loading, was analysed at similar $W_{avg}$ values of 0.16 and 0.19 mm (Arrows in Fig. 2a), as shown in Fig. 3. In this figure, the drag forces measured at each CrAl coating layer showed a strong dependency on the negative substrate bias. This result demonstrated that the resistance to plastic deformation of the CrAl coating layer is expected to be proportional to the applied negative substrate bias. Regardless of the different scratch traces at each CrAl coating layer, no gross spallation or tensile cracks of the coating layer were found in the entire stroke length up to the applied normal load of 50 N.

### 3.2 Wear behaviours

Fig. 4a shows the wear volume results of CrAl-coated claddings and the reference Zr cladding as a function of the number of cycles. The wear volume of the CrAl-coated claddings are significantly less than that of the Zr cladding regardless of the applied negative substrate bias. No significant difference was found at up to $10^6$ cycles even though the transition cycles are dependent on the applied negative substrate bias. The variation of wear depth also showed a similar behaviour with that of the wear volume, as shown in Fig. 4b. However, a higher maximum wear depth appeared under A-100V conditions, which has the thinnest coating thickness of 10 $\mu$m. This layer is removed by interacting with the Zr-based grid spring under the fretting condition, and the wear damages are accelerated on Zr cladding substrate owing to the relatively lower hardness. Thus, a coating thickness of 10 $\mu$m under the A-100V condition is insufficient to prevent fretting wear damage used in this test condition. These results demonstrated that if the coating layer thickness is 30 $\mu$m or more, the CrAl-coating layer deposited with a negative substrate bias from -100 to -250V, exhibits better wear-resistant properties than the current Zr-based cladding when the current Zr-based grid spring is applied as a support material.




(a) Wear volume (b) Maximum wear depth

Fig 4. Variation of wear volume and maximum wear depth at each Cr-coated cladding and a commercial Zr-based alloy

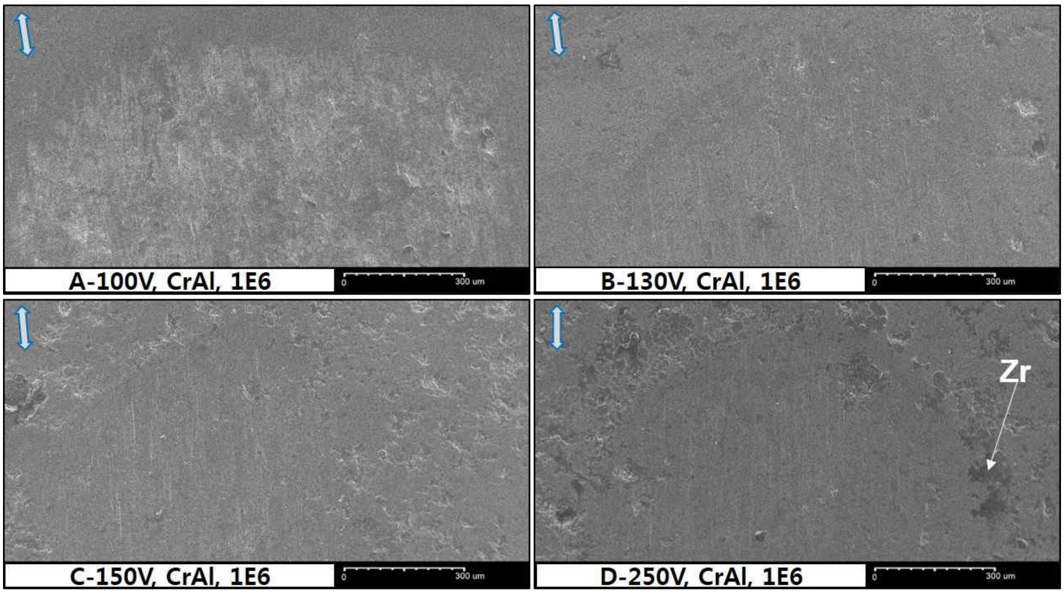



Fig 5. Typical SEM micrographs of CrAl-coated claddings at upper boundary of worn surface. (Note-Arrow indicates fretting direction)

### 3.3 Surface wear features

Fig. 5 shows the fretting wear mechanism at the upper boundary of the worn surface at each CrAl-coated cladding at a normal force of 10 N, slip amplitude of 100 μm, and fretting cycles of $1x10^6$. The worn surface under A-100V condition shows that the formation of a severe deformation layer and its crack propagation are easily observed, indicating that the wear debris is detached and partially transferred to the grid spring by an adhesive wear mechanism. In the case of B-130V, C-150V, and D-250V conditions with thicker coating layers, however, deformation layers are well developed on the worn surface of CrAl coating, acting as load-bearing layers for restricting the formation of cracks and wear debris. In addition, it is expected that the coating layer deposited by a higher negative substrate bias needs more cycles for accumulating the fretting wear damage.

Based on the energy dispersive spectroscopy (EDS) analysis of the worn surface of a grid spring, the dark colours in Fig. 6 turned out to be Cr elements with a negligible amount of Al elements transferred from the CrAl coating layer. This result indicates that the higher the negative substrate bias is, the stronger the tendency of adhesive wear, providing strong evidence of a lower wear depth increase under D-250V conditions.

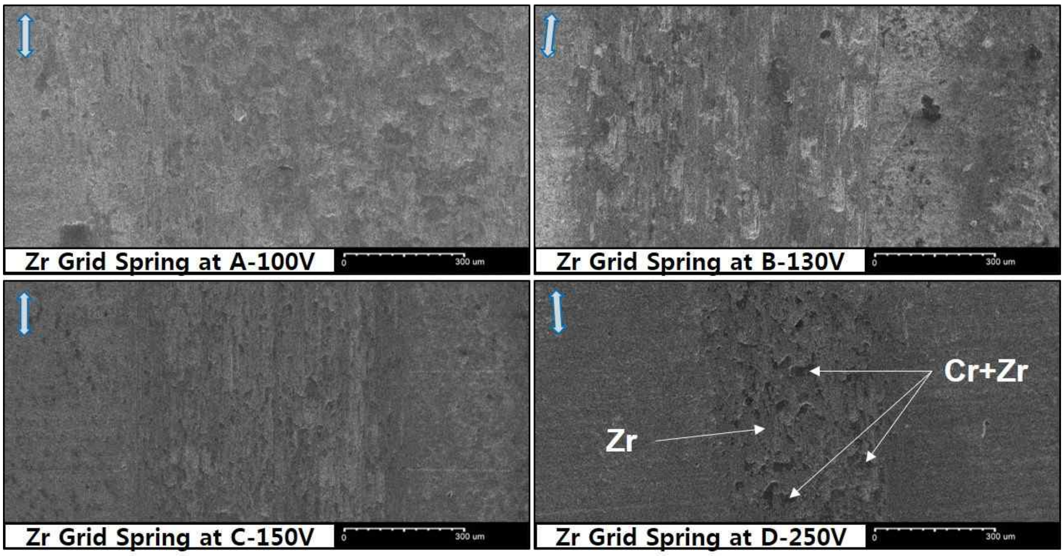



Fig 6. Wear scar morphologies of Zr-based grid spring against CrAl-coated claddings. (Note-Arrow indicates fretting direction)

## 4. Summary

Four kinds of CrAl-coated claddings with a different negative substrate bias have been submitted to the constant load scratch test and fretting wear test, focusing on the deformation behaviour, adhesion strength, and fretting wear mechanism of the coating layer. No gross spallation and tensile cracks in CrAl-coated claddings were found in the entire stroke length up to the applied normal load of 50 N. However, the final coating thickness should be determined when considering the non-uniform mechanical properties within the coating layer. Fretting wear results show that CrAl-coating cladding exhibits better wear-resistant properties than current Zr-based cladding when a current Zr-based grid spring is applied as the support materials. The dominant wear mechanism in room temperature water is adhesive wear by material transfer between the CrAl coating and Zr-based grid spring.

## 5. Acknowledgement

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