PennState College of Earth and Mineral Sciences

Department of Materials Science and Engineering

Advanced Accident-Tolerant Ceramic Coatings for Nuclear Fuel Cladding

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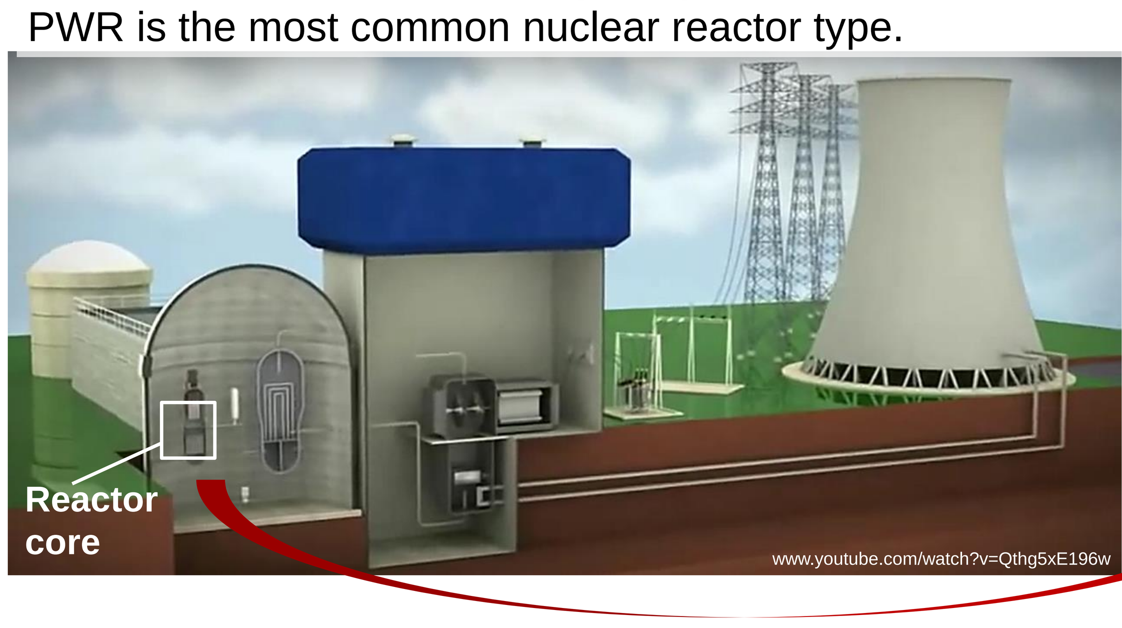
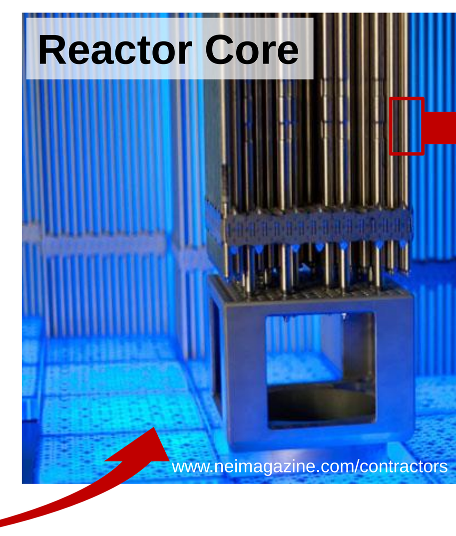
Introduction

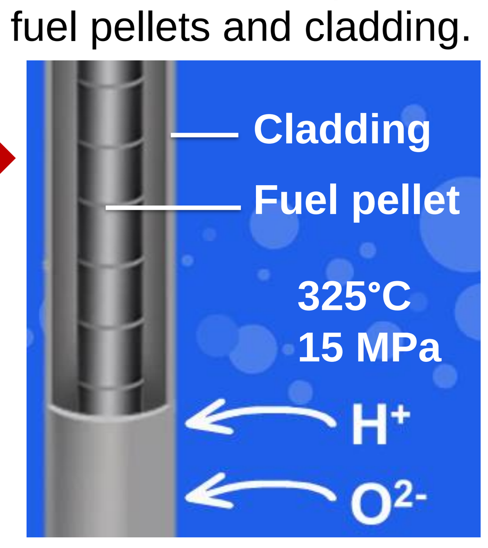
Pressurized Water Reactor (PWR)
PWR is the most common nuclear reactor type.





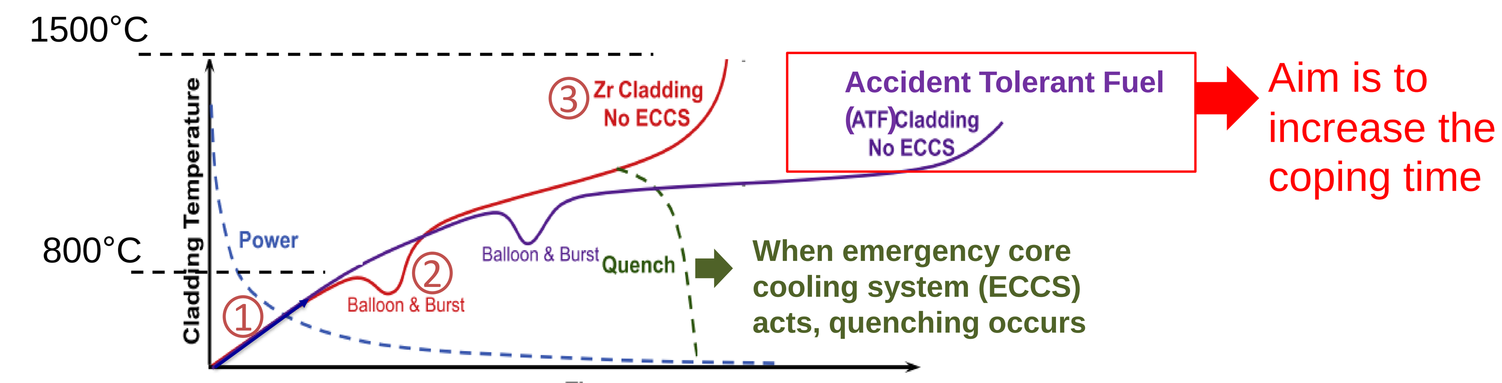
Energy is created in the reactor core using 4 m long fuel rods.

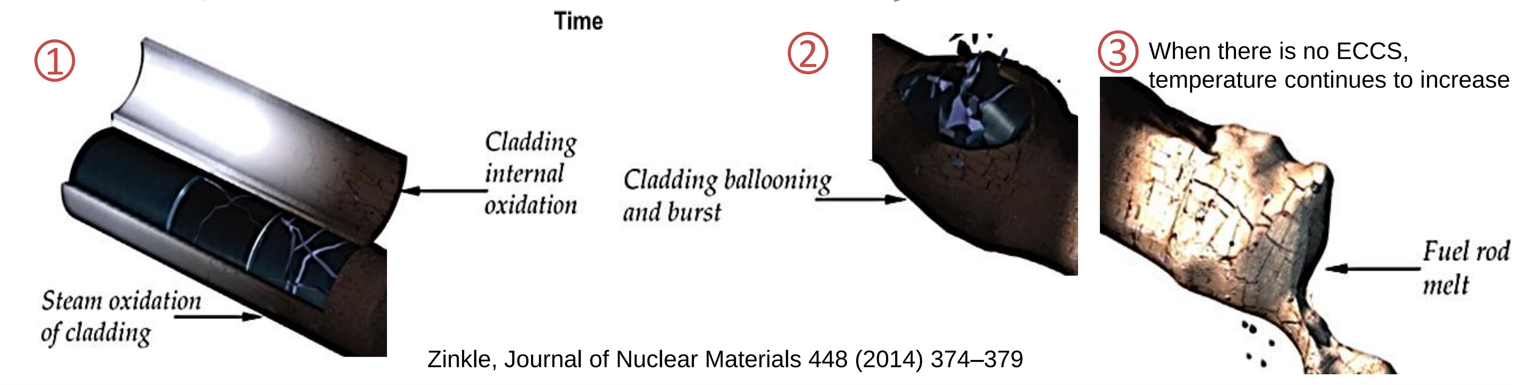
A fuel rod is composed of fuel pellets and cladding.



At normal operation:
Coolant water surrounding the fuel rod at 325°C and 15.5 MPa

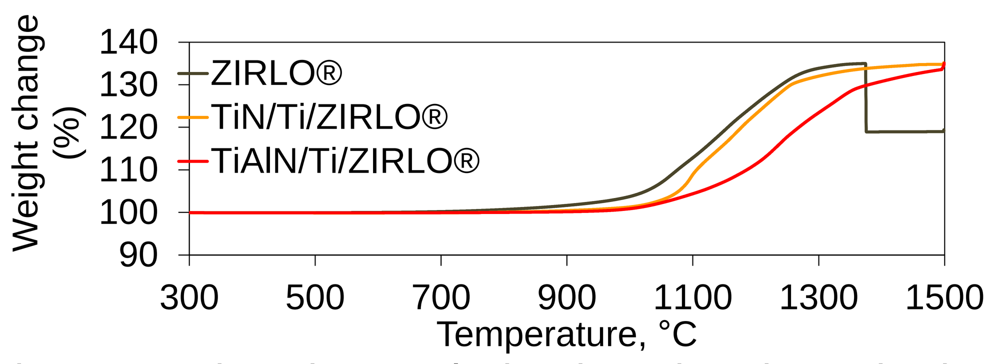
However, in case of a loss-of-coolant accident, cladding degradation **motivated** research for **Accident Tolerant Fuels** (ATF).



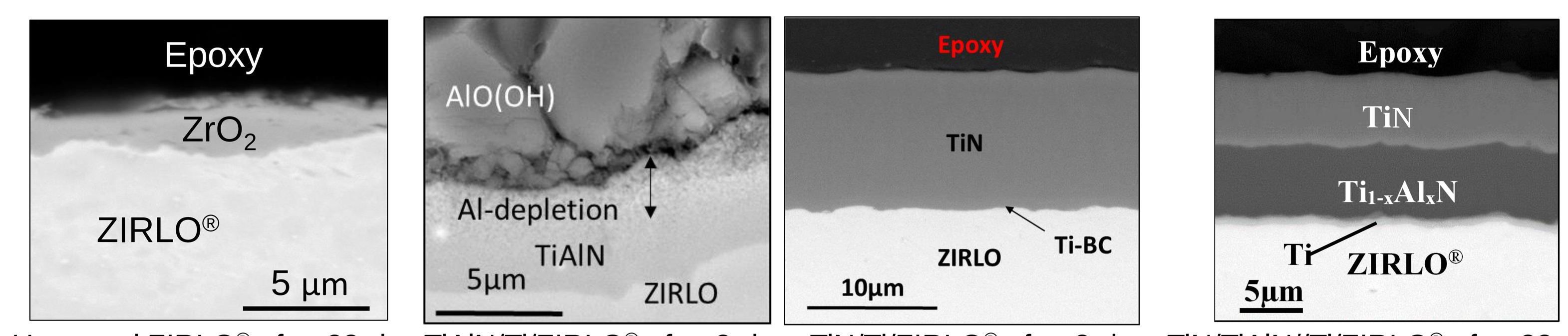


Zinkle, Journal of Nuclear Materials 448 (2014) 374–379

Background



Thermogravimetric analysis in air showed the oxidation onset temperature delay in coated samples.



Uncoated ZIRLO® after 23-d | TiAlN/Ti/ZIRLO® after 3-d | TiN/Ti/ZIRLO® after 3-d | TiN/TiAlN//Ti/ZIRLO® after 33- d

Autoclave test in static pure water at 360°C and 18.7 MPa showed oxidation behavior of uncoated and coated samples.

TiN and TiAlN coating application on zirconium-based alloys delay oxidation initiation temperature and prevent substrate corrosion.

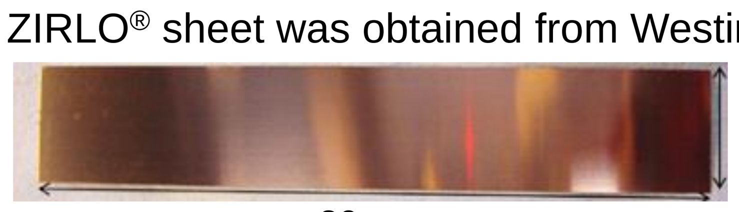
TiN and TiAlN coating is a promising ATF concept.

Further study is necessary to enhance coating adhesion.

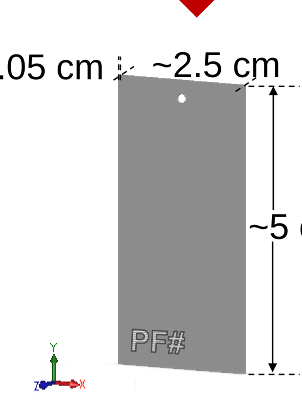
Alat et al., Surf. Coat. Technol. 281 (2015) 133-143
Alat et al., J. Nucl. Materials 478 (2016) 236-244.

Experimental Method

ZIRLO® sheet was obtained from Westinghouse

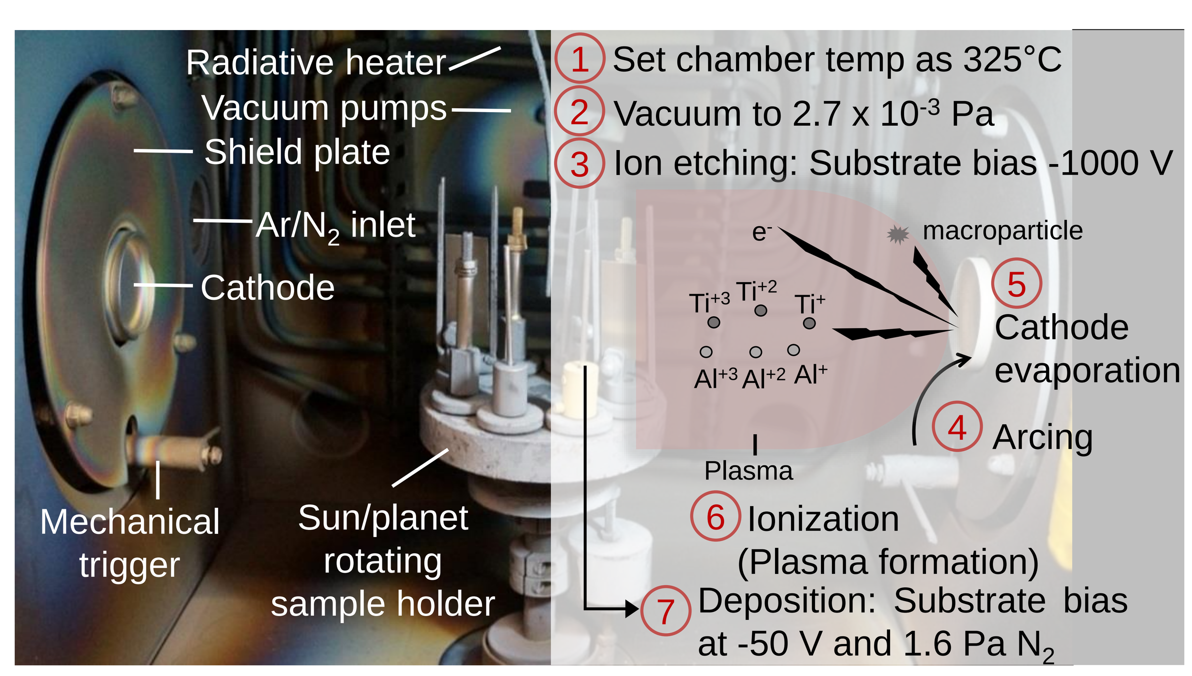


Cutting with diamond

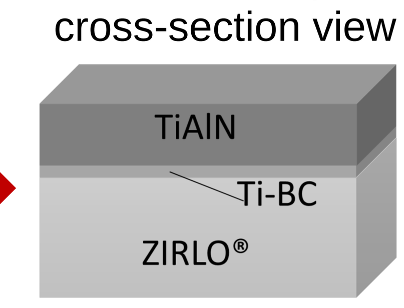


Surface grinding to remove oxides and get 0.25 µm R_a surface roughness

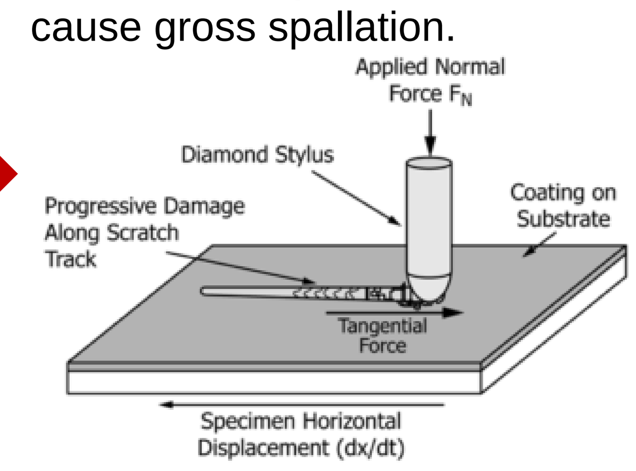
Cathodic arc physical vapor deposition (CA-PVD)



Coated sample cross-section view



Coating adhesion to substrate evaluation:
Scratch testing to determine critical load to cause gross spallation.



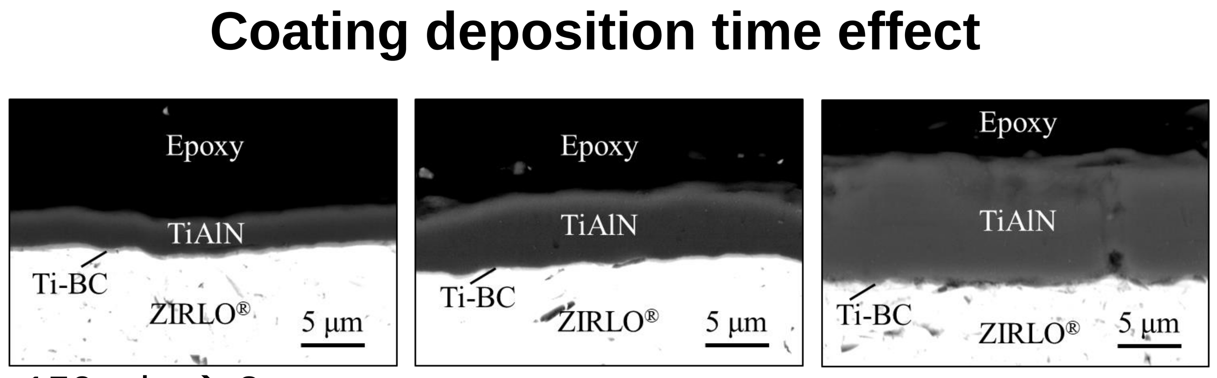
Parameters:
1 cm scratch in 1 minute up to 50 N load

Grazing incidence X-ray diffraction (GIXRD) for phase identification
Scanning electron microscopy for high-resolution imaging

Results and Discussion

Coating deposition time effect

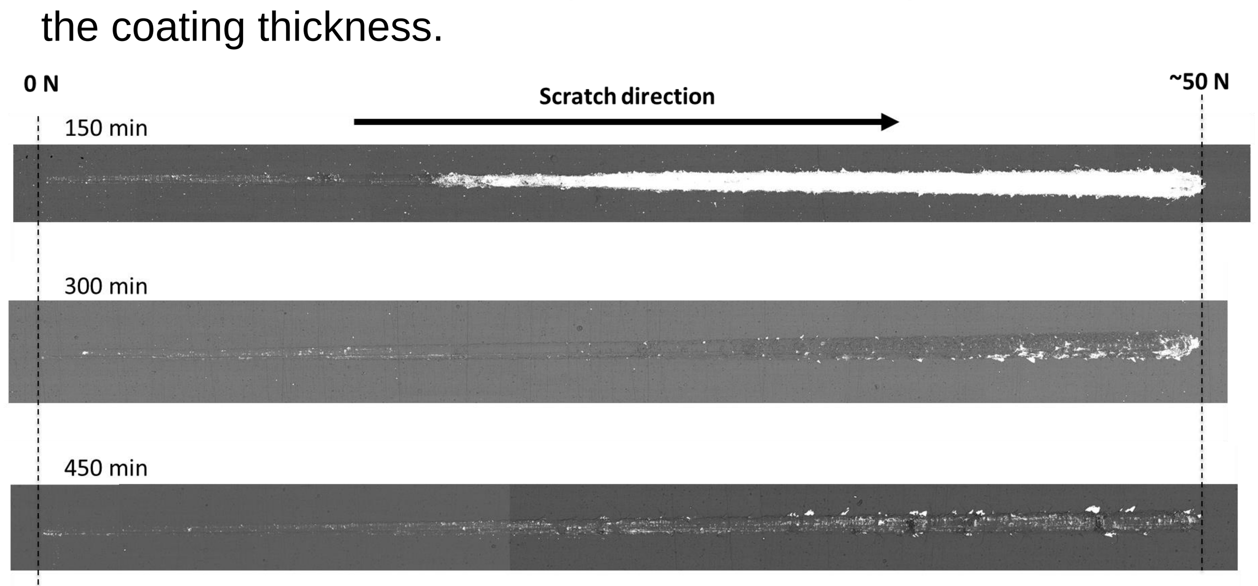
As-deposited coating characterization



150 min → 2 µm | 300 min → ~4 µm | 450 min → ~10 µm

There is a linear relationship between the deposition time and the coating thickness.

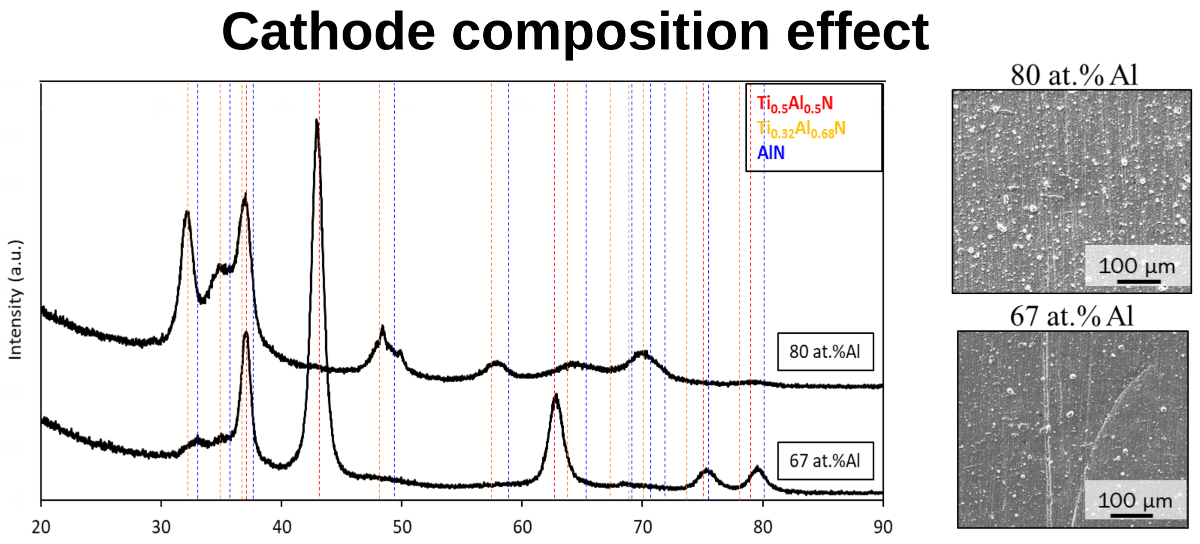
Scratch testing results



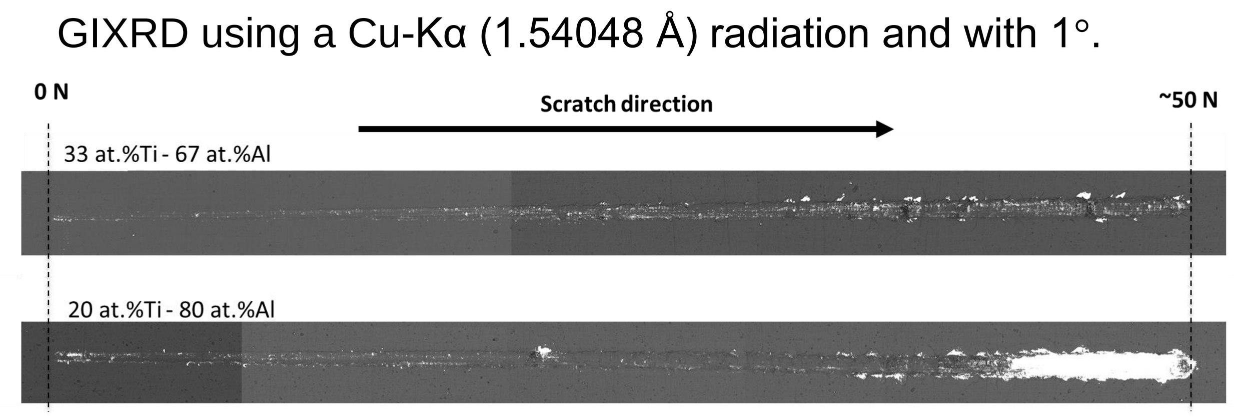
Coating thickness ↑ leads to adhesion ↑

Better mechanical performance with thicker coatings is due to a combination of high load-bearing capability of the thicker hard coating and ductile substrate deformation prior to the coating spallation.

Cathode composition effect



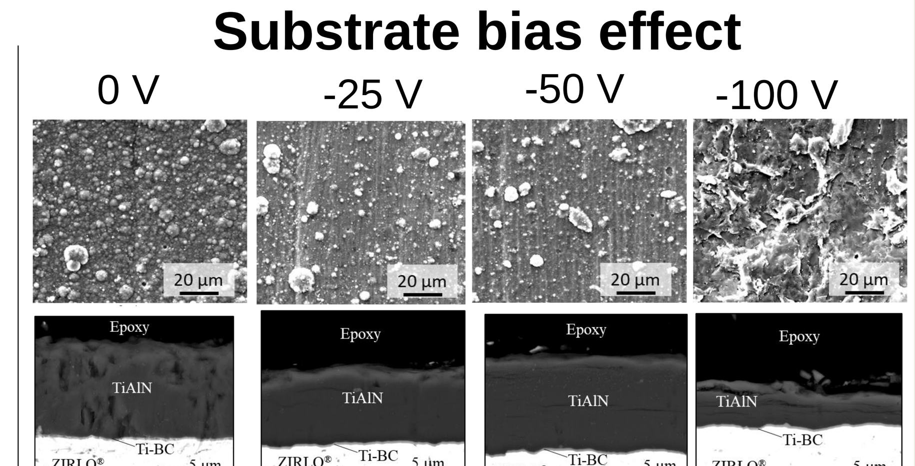
GIXRD using a Cu-Kα (1.54048 Å) radiation and with 1°.



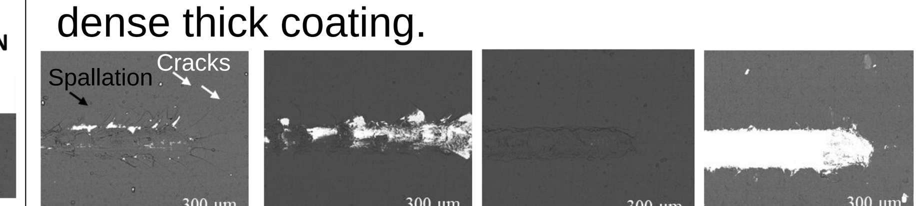
33 at.%Ti-67 at.%Al provides better adhesion

For a cathode composition of 67 at.% Al, the coating phase is determined to be $Ti_{0.5}Al_{0.5}N$ (cubic). For 80 at.% Al, peak patterns were fitting with the $Ti_{0.32}Al_{0.68}N$ phase and AlN (hexagonal). Increased aluminum content led to macroparticle increase, which decreases coating adhesion.

Substrate bias effect



0 V substrate bias resulted in macroparticle covered surface with porous coating. 100 V lead to nonuniform coating. 50 V provided dense thick coating.



- 50 V is the optimum substrate bias.

The higher critical load required for gross spallation in the case of 50 V substrate bias can be attributed to ejection of metal ions from the surface or penetration to the substrate lattice during ion bombardment leading to surface roughness that can enhance coating adhesion

Conclusions

- Coating thickness increase results in critical load increase for gross spallation due to complex stress state and increase in thickness leads to increase in adhesion.
- Coating composition mirrors the cathode composition. In the case of lower aluminum content having cathode (33 at.%Ti-67 at.%Al), due to the lower aluminum content in the coating, cubic crystal structure is observed which provides higher mechanical performance and lower amount of macroparticles enhancing the coating adhesion.
- There is an optimum substrate bias value of -50 V. Increasing substrate bias decreases macroparticles with enhanced coating density up to a critical value but decreases coating uniformity afterwards.

Acknowledgements

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